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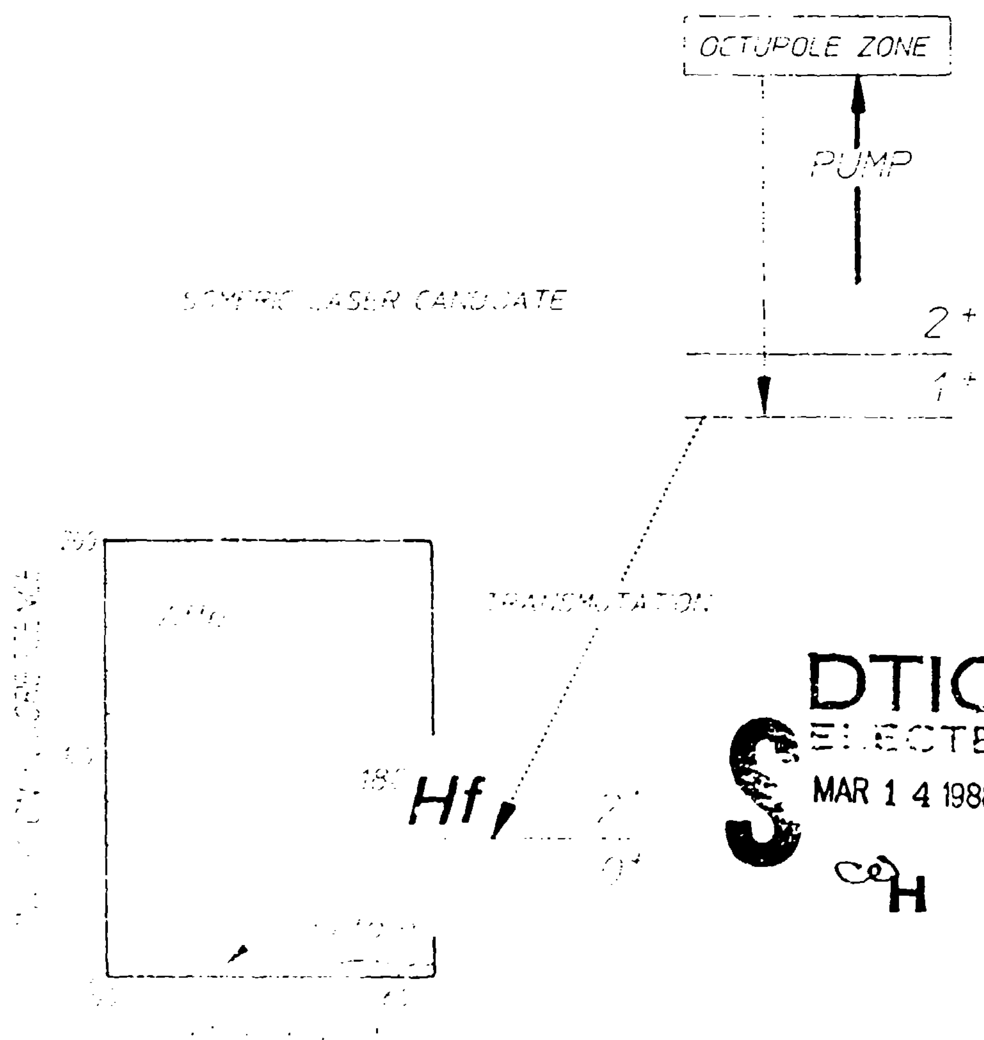

Report GRL/8703

PROOF OF THE FEASIBILITY OF COHERENT AND INCOHERENT SCHEMES FOR PUMPING A GAMMA-RAY LASER

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Recent approaches to the problem of the gamma-ray laser have focused upon upconversion techniques in which metastable nuclei are pumped with long wavelength radiation. At the nuclear level the storage of energy can approach tera-Joules (10^{12} J) per liter for thousands of years. However, any plan to use such a resource for a gamma-ray laser poses problems of a broad interdisciplinary nature requiring the fusion of concepts taken (continued next page)

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20. Abstract (continued)

from relatively unrelated fields of physics. Our research group has described several means through which this energy might be coupled to the radiation fields with cross sections for stimulated emission that could reach 10^{-17} cm^2. Such a stimulated release could lead to output powers as great as 3×10^{21} Watts/liter. Since 1978 we have pursued an approach for the upconversion of longer wavelength radiation incident upon isomeric nuclear populations that can avoid many of the difficulties encountered with traditional concepts of single photon pumping. Recent experiments have confirmed the general feasibility and have indicated that a gamma-ray laser is feasible if the right combination of energy levels and branching ratios exists in some real material. Of the 1886 distinguishable nuclear materials, the present state-of-the-art has been adequate to identify 29 first-class candidates, but further evaluation cannot proceed without remeasurements of nuclear properties with higher precision. A laser-grade datebase of nuclear properties does not yet exist, but the techniques for constructing one have been developed under this contract and are now being utilized. Resolution of the question of the feasibility of a gamma-ray laser now rests upon the determination of: 1) the identity of the best candidate, 2) the threshold level of laser output, and 3) the upconversion driver for that material.

This quarter's report focuses upon continued development of one of the new technologies for the screening of the laser candidates. It is the nuclear analog of the optical double resonance methods which produced much of the database at the molecular level that was of such essential use in the development of conventional lasers. Applied most recently to the study of levels which might be used in dumping isomeric populations into freely radiating states, it produced an unexpected result of major importance. In several test isotopes, a class of extremely useful states was discovered that could radiatively couple to both normal and isomeric states of a nucleus spanning large changes of angular momentum.

The achievements of this quarter culminated in the major milestone demonstration of the dumping of population of the first ~~of the~~ 29 actual candidates for a gamma-ray laser. One of the least attractive of the 29 isomers because of a large change in angular momentum of $8\hbar$, $^{180}Ta^m$ was the only one of the candidates available in milligram quantities. Pumped with the bremsstrahlung from a medical linear accelerator having its maximum output near 2 MeV, an astonishingly large cross section of 40,000 was found on the scale where unity results in a large yield. This corresponds to a partial width for the useful pumping of this isomer of 0.5 eV, a value nearly 10^6 in excess of the rule-of-thumb supposed to limit the useful absorption widths to 1 μeV for the pumping of nuclei with flash x-rays. The extent of the success with this relatively poor candidate considerably strengthens the probabilities that a more suitable isomer for a gamma-ray laser exists among the better candidates.

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PREFACE

The nuclear analog of the ruby laser embodies the simplest concepts for a gamma-ray laser. Not surprisingly, the greatest rate of achievement in the quest for a subAngstrom laser continues in that direction. This quarterly report focuses upon the second major milestone achieved in 1987. The first had shown that bandwidth funneling works at the nuclear level, just as it did for ruby on the molecular scale. Experiments pumping ^{77}Se and ^{79}Br produced eleven orders of magnitude increase in fluorescence intensity over what could have been obtained by direct excitation. Now the second milestone has demonstrated great success in optically pumping the first of the 29 actual candidates for a gamma-ray laser, $^{180}Ta^{m}$.

Not a particularly attractive candidate, <u>a priori</u>, $^{180}Ta^{m}$ was the only one for which a macroscopic sample was available. The need to span a formidably large $\Delta J = 8$ between isomer and fluorescence level supported little initial enthusiasm for this nucleus. When actually pumped, however, it showed the largest integrated cross section ever reported for interband transfer in any material, 4×10^{-22} cm^2 eV. This is an enormous value for bandwidth funneling, being about 10,000 times greater than what is characteristic of an efficient nucleus such as ^{77}Se or ^{79}Br which starts in the ground state and transfers to the isomer. The optical pumping of an isomer itself had never been previously reported, so nothing was available for a more direct comparison with the present result.

The partial width for absorption from $^{180}Ta^{m}$ isomer to fluorescence was measured to be about 0.[illegible] eV, a value far exceeding the 1 meV usually offered as a rule of thumb that would limit the interband transfer of nuclear population. Now, the first attempts to extend these results to

other species have demonstrated widths approaching 0.1 eV for the excitation of isomers in five other nuclei.

Perhaps, as suggested last quarter, collective oscillations which break the symmetries of the nuclei provide this major windfall making it easier to dump isomers by mixing single particle states needed in the transfer process. Much more experimentation will be needed to identify whether this is the actual mechanism responsible and to understand if the lessons taught by $^{180}Ta^{m}$ are generally applicable in the pool of candidate isomers. Now, such experiments are facilitated by the widths themselves, which have reduced the level of effort to practical dimensions. The experiments described in this report show clear fluorescence signals can be obtained on the current scale of illuminating milligrams of material with intensities which peak in time at only a few Watts/cm^2, even when integrated over all wavelengths. At this level, meaningful experiments can be performed on the next three candidate isomers when samples become physically available.

Continuing the preparation of this report as an "in-house" journal, this series presents material to reflect the individual contributions of the teams of research faculty and graduate students involved in these phases of the research. In this regard I wish to thank all our staff for their splendid efforts in supporting the preparation of these manuscripts to a rather demanding timetable.

- C. B. Collins
- Director
- Center for Quantum Electronics

MAJOR MILESTONE REPORT

Affecting the Feasibility of Coherent and Incoherent Schemes
for Pumping a Gamma-Ray Laser

October 16, 1987

C. B. Collins, Center for Quantum Electronics, University of Texas at Dallas

Achievement

X-ray pulses have been used to pump very large amounts of nuclear fluorescence from hundred-microgram quantities of the first of the 29 candidate isomers to be tested for a gamma-ray laser.

Technical Background

The nuclear analog of the ruby laser embodies the simplest concepts for a gamma-ray laser. Not surprisingly, the greatest rate of achievement in the quest for a subAngstrom laser has developed in that direction.

For ruby the identification and exploitation of a bandwidth funnel were the critical keys in the development of the first laser. There was a broad absorption band linked through efficient cascading to the narrow laser level.

Nuclei to be used in the analog of the ruby laser can start in either ground or isomeric states. However, with the latter, most of the output power can be derived from the energy stored in the isomeric state at its creation. Then in addition to the obvious need to transfer energy in order to reach a fluorescence level to be populated for lasing, there must also be a substantial transfer of angular momentum. Major milestones we reported previously proved that bandwidth funneling worked for nuclei of simulated candidates but the change in angular momenta did not need to be very large in those materials. It was found that nuclear fluorescence could be pumped by flash x-rays through integrated cross-sections as large as 30 in the usual units ($\times 10^{-29}$ cm^2 keV). However, many of the 29 actual candidate isomers have angular momenta which are very different from the laser levels to which they are supposed to be dumped. The concern has lingered that actual candidates would have pumping cross-sections of only a minute fraction of a unit, if not actually zero, and so would be entirely useless.

Reported here is an experimental breakthrough which answers this concern. *A population of candidate isomer ^{180m}Ta has been dumped through an integrated cross-section of 80,000 units.*

Report

Most of the nuclear data bases do not yet record the recent discovery that nature's rarest element, tantalum-180, is actually an isomer lying 80 keV above a ground state which is unstable against transmutation to tungsten and hafnium. Having a very high spin of 9^-, the isomer ^{180m}Ta has a cosmic lifetime and traces remain on earth mixed with the normal commercial tantalum, ^{181}Ta. Because of a curious importance to the cosmic nucleosynthesis of the heavier elements, astrophysicists have studied the energetics of the lower levels of the ^{180}Ta system as shown in Fig. 1, except for the broad level near 2000 keV which has been added here as a result of our work.

In this major milestone experiment about 500 μg of naturally occuring isomeric ^{180m}Ta diluted in 4.75 g of ^{181}Ta were irradiated with the bremsstrahlung from a 6 MeV linac. The accumulated dose at 2 MeV near the peak of the spectrum was 3.8×10^{10} photons/cm^2/keV. The excitation energy of the gateway state shown in Fig. 1 was assumed to be 2000 keV in order to obtain a minimum value for the cross section, since smaller fluxes were available at even higher energies.

The spectrum of the radioactive debris shown in Fig. 2 demonstrates that isomeric ^{180m}Ta nuclei were pumped through a broad level cascading finally to the unstable ground state of ^{180}Ta. The amount of debris determines the cross section for the process to be about 80,000 of the usual units ($\times 10^{-29}$ cm^2 keV).

Significance

There is a threefold significance to this demonstration of the efficiency for pumping isomers with x-rays.

1) The first real isomer to be tested for a gamma-ray laser was successfully pumped down with an astonishingly large cross section of 80,000 on a scale where 10 describes a fully allowed process.

2) The nuclear analog to the ruby laser is a fully viable scheme for a gamma-ray laser, and ^{180m}Ta narrowly misses being an acceptable candidate. It performed about 10^4 times better than would have been expected theoretically.

3) These results with a seemingly unattractive candidate indicate the probabilities should be raised for full success of one of the other 28 materials.

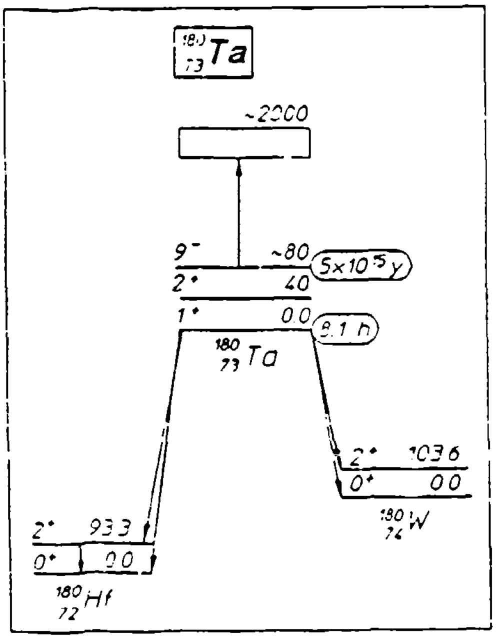

Figure 1: Schematic energy level diagram of ^{180}Ta and its daughters. Half-lives are shown in ovals to the right of the ground and isomeric levels. Energies are in keV. The pump band is shown by the arrow pointing upward to the broad state represented by the rectangle. Cascade through the potential laser levels of ^{180}Ta is not known, but leads finally to the ground state. Electron capture to the left and beta decay to the right are indicated by the diagonal downward arrows. The final debris from pumping down the isomer is found in the fluorescence from the 93.3 keV transition of ^{180}Hf characterized by the 8.1 hour lifetime of its ^{180}Ta parent.

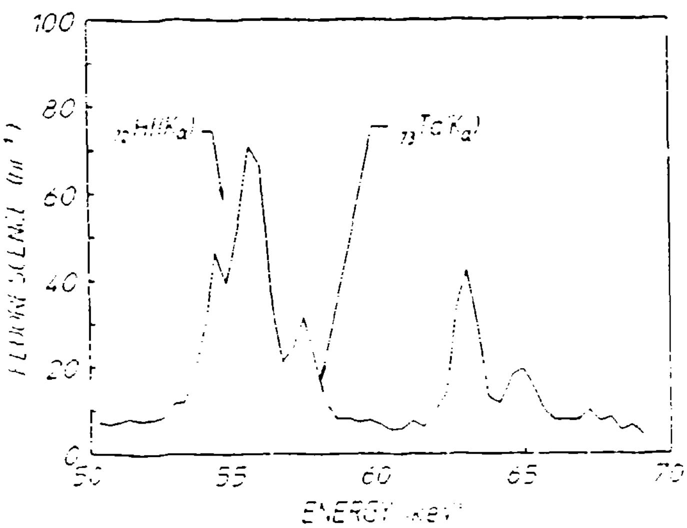

Figure 2: Dotted and solid curves show, respectively, the spectra obtained before and after dumping some of the 500 μg of isomeric ^{180m}Ta. An HPGe detector was used to obtain the dotted spectrum excited in the 4.75 g of diluent ^{181}Ta by the traces of natural activity in the counting shield. The solid curve shows activity resulting from the transmutation of the pumped ^{180}Ta measured in the same sample and counting system after irradiation. The prominent addition is the K_α pair from hafnium excited by the internal conversion of the 93 keV transition shown in Fig. 1.

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THE GAMMA-RAY LASER - STATUS AND ISSUES FOR 1988

by C. B. Collins

Efforts to demonstrate the feasibility of a gamma-ray laser scored major advances in 1987. Culminating with the successes in optically pumping the first of the 29 actual candidate isomers, priority issues were brought into better focus by the lessons learned from a wealth of new results. Perceptions were advanced so greatly that it has become necessary to reassess the critical issues remaining for 1988. Only the bottom line remains the same. *A gamma-ray laser is feasible if the right combination of energy levels occurs in some real material.* The likelihood of this favorable arrangement has been markedly increased by the experimental results of 1987.

From the inception of the gamma-ray laser program, it had been realized that levels of nuclear excitation which might be efficiently stimulated in a gamma-ray laser would be very difficult to pump directly. To have sharply-peaked cross sections for stimulated emission, such levels must have very narrow widths for interaction with the radiation field. This is a fundamental attribute that had led to the facile criticism that "absorption widths in nuclei are too narrow to permit effective pumping with x-rays."

The same concerns had been voiced in atomic physics before Maiman's great discovery, and it has proven very useful to pursue this analogy between ruby and gamma-ray lasers. The identification and exploitation of a bandwidth funnel in ruby were the critical keys in the development of the first laser. There was a broad absorption band exciting a state of Cr^{3+} which quickly decayed by cascading its population into levels of lower energy. A reasonably favorable pattern of branching insured that much of the cascading populated the narrow level. At the core of our simplest proposal[1] for pumping a gamma-ray laser is the use of the analog of this effect at the nuclear level as shown in Fig. 1. A detailed analysis of this mechanism was reviewed as early[2] as 1982 and has been emphasized in more recent reports[3,4] together with the breakthrough actually demonstrating the great utility of bandwidth funneling at the nuclear level. Yields of gamma-ray fluorescence in ^{77}Se and ^{79}Br were enhanced by eleven orders of magnitude by this effect.[5]

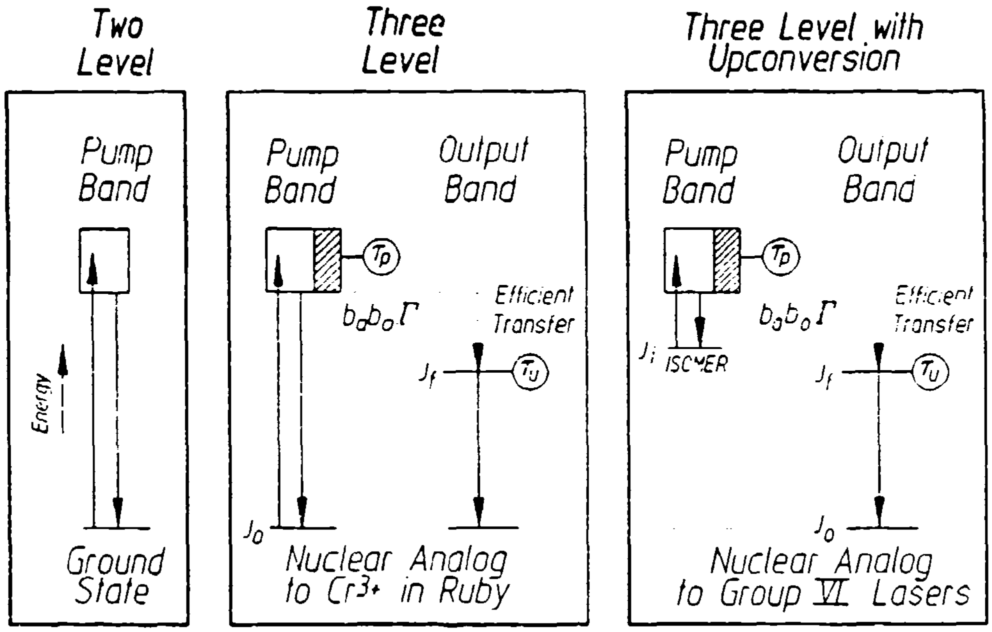

Figure 1: Schematic representation of the energetics of the priority schemes for pumping a gamma-ray laser with flash x-rays. The large width of the level defining the pump band is implied by the height of the rectangle representing the level and the shaded portion indicates that fraction, b_0, which is attributed to the transition to the upper laser level. Angular momenta of the ground, isomeric, and fluorescent levels are denoted by J_0, J_i, and J_f, respectively.
(a) Traditional two level approach.
(b) Three-level analog of the ruby laser serving to illustrate the important concept of bandwidth funneling.
(c) Refinement of the three-level scheme which incorporates upconversion in order to lessen the energy per photon which must be supplied in the pumping step.

Also shown in Fig. 1 is a further refinement of the incoherent pumping scheme benefiting from upconversion. As has been often discussed,[2,6] upconversion as shown in Fig. 1c has many advantages. Most prior reports have emphasized those tending to enhance performance and efficiencies; however, upconversion also makes threshold itself much more accessible. Higher energy isomers need less pump energy to reach the broad states that would optimize bandwidth funneling, and the required pump energies can fall in the range where strong x-ray lines may be found to concentrate the spectral intensity.

Whether or not the initial state being pumped is isomeric, the principal figure of merit for bandwidth funneling is the partial width for the transfer, $b_a b_o \Gamma$. Constituent parameters are identified in Fig. 2 where it can be seen that the branching ratios b_a and b_o specify the probabilities that a population pumped by absorption into the i-th broad level will decay back into the initial or fluorescent levels, respectively. It is not often that the sum of branching ratios is unity, as channels of decay to other levels are likely. However, the maximum value of partial width for a particular level i occurs when $b_a = b_o = 0.5$.

In 1986 one of the strongest tenets of theoretical dogma insisted that for processes of optical pumping involving long-lived isomers,

$$b_a b_o \Gamma \leq 1.0\ \mu eV \qquad , \tag{1}$$

so that the efficacy of bandwidth funneling would be seriously limited in all important cases. The first major milestone[5,7,8] of 1987 demonstrated partial widths of 39, 5, and 94 μeV for the excitation with bremsstrahlung of isomers of ^{77}Se, ^{79}Br, and ^{115}In, respectively, from ground state populations. While providing a "moral victory" by breaking the absolute limits of Eq. (1), these results still left an aura of credibility to the rule-of-thumb that partial widths for isomers would be limited to the order of magnitude of μeV.

The actual measurement of partial widths involves the correlation of fluorescence yields excited by a pulse of continuous x-rays in the scheme of Fig. 2 with those expected from the expression,[4-9]

$$N_f = N_o \sum_i f_i \frac{\phi_i}{A} \qquad , \tag{2a}$$

where N_o and N_f are the numbers of initial and fluorescent nuclei respectively, (ϕ_i/A) is the spectral intensity of the bremsstrahlung in keV/keV/cm^2 at the energy E_i of the i-th pump band, and the summation is taken over all of the possible pump bands capable of cascading to the same fluorescence level of interest. The f_i is a combination of nuclear parameters including the partial width $b_a b_o \Gamma$ in keV,

$$f_i = \frac{(\pi b_a b_o \Gamma \sigma_o / 2)_i}{E_i} \qquad , \tag{2b}$$

where σ_o is the peak o. the Breit-Wigner cross section for the absorption step. The combination of parameters in the numerator of Eq. (2b) is termed the integrated cross section for the transfer of population according to the scheme of Fig. 2.

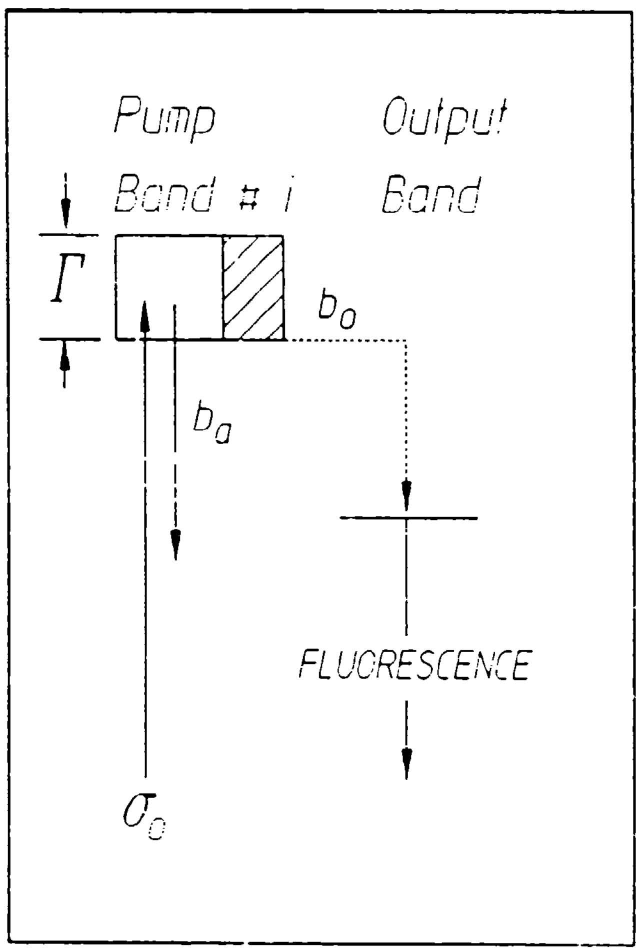

Figure 2: Schematic representation of the decay modes of a gateway state of width Γ sufficiently large to promote bandwidth funneling. The initial state from which population is excited with an absorption cross section σ_o can be either ground or isomeric.

Of the many potential systems for a test of the formulations of Eq.(2a) and (2b), the literature[10] supports the calculation of integrated cross sections for very few. Table I summarized those which are known with sufficient accuracy to serve as standards. In the convenient units of 10^{-29} cm^2 keV, values range from the order of unity to a few tens for bandwidth funnels that are sufficient for demonstrations of nuclear fluorescence from reasonable amounts of material at readily accessible levels of input. The largest integrated cross section ever inferred[11] for transfer to an isomer was 380 ($\times 10^{-29}$ cm^2 keV) for a pump band in ^{87}Sr at 2.66 MeV. Being of singular size, it was not considered as a proof of the fallibility of the rule of Eq. (1), established some years after that report.

Table I

Summary of nuclides, pump lines, and integrated cross sections for the excitation of delayed fluorescence suitable for use as calibration standards.

	PUMP LINE keV	$\pi b_a b_o \Gamma \sigma_o/2$ 10^{-29} cm^2 keV
^{79}Br	761	6.2
^{77}Se	250	0.20
	480	0.87
	818	0.7
	1005	30
^{115}In	1078	20

In a series of experiments[8,12] we conducted in 1987 that were designed to confirm the optical pumping model of Eqs.(2a) and (2b), samples of the standard nuclei of Table I were pumped with intense pulses of bremsstrahlung from the DNA nuclear simulator, PITHON. The clear signal-to-noise ratios that typified subsequent measurements of nuclear fluorescence excited through the pump bands of Table I are shown in Fig. 3. The quality of such data enabled us to "invert" Eqs. (2a) and (2b) so that the spectral intensities of the pump could be obtained at three energies from the measured values of fluorescence excited from a single pulse. Figure 4 shows a typical result[3] in comparison with a

calculation of the bremsstrahlung spectrum from that particular source on that particular shot. Both measurement and calculation are absolutes with no free parameters to adjust. Such a direct measurement of the spectrum from an intense pulse of x-ray continua had not been previously reported, and the agreement with expectations was gratifying. Moreover, it confirmed that this type of nuclear analog of the optical double resonance measurements at the atomic level can be performed with a reasonable level of accuracy.

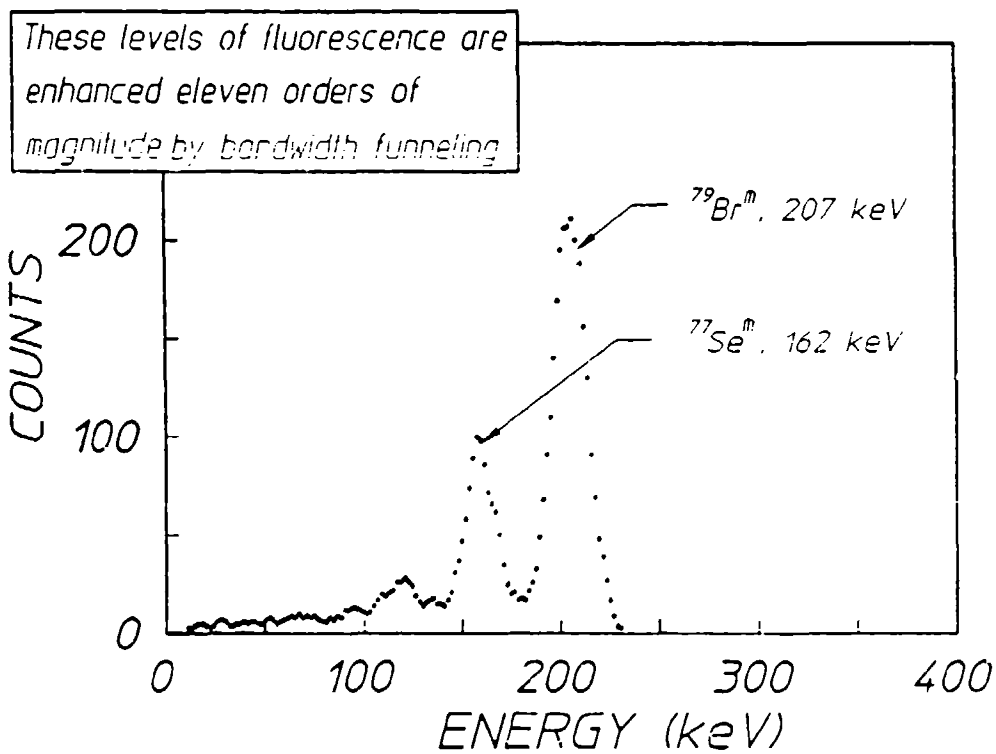

Figure 3: Fluorescence spectrum from a target containing 1.25 g LiBr and 1.20 g of elemental Se, both in natural abundances, excited with a single irradiation by the bremsstrahlung produced by the DNA/PITHON electron beam device. Acquisition time of this data was 80 s. Prominent lines are contributed by the isomeric transitions indicated.

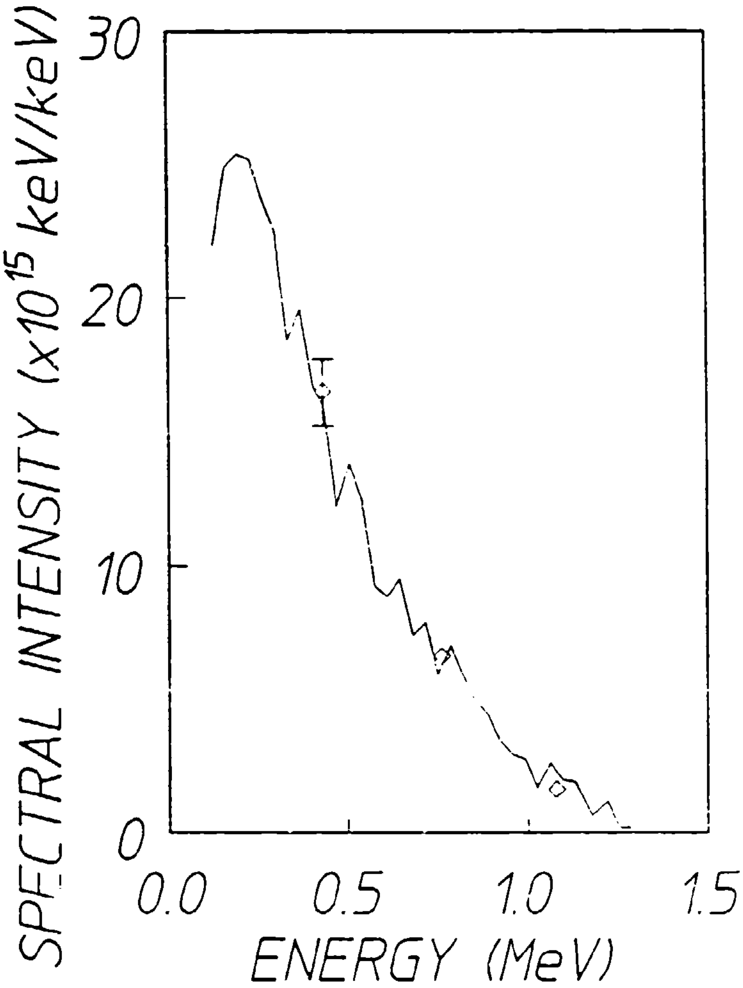

Figure 4: Data points plot the spectral intensities measured directly with our nuclear activation technique using the parameters of Table I in comparison to the spectrum computed with a coupled electron, photon transport code for a typical PITHON shot. Vertical bars show uncertainty in the measurement at the one point for which that uncertainty was larger than the plotted size of the symbol.

Tempering expectations that these successes might be readily extended to the pumping of actual isomeric candidates for a gamma-ray laser was a concern for the conservation of various projections of the angular momenta of the nuclei. Many of the interesting isomers belong to the class of nuclei deformed from the normally spherical shape. For those systems there is a quantum number of dominant importance, K, which is the projection of individual nucleonic angular momenta upon the axis of elongation. To this is added the collective rotation of the nucleus to obtain the total angular momentum J. The resulting system of energy levels resembles those of a diatomic molecule for which

$$E_x(K,J) = E_x(K) + B_x J(J+1) \quad , \qquad (3)$$

where $J \geq K \geq 0$ and J takes values $|K|$, $|K| + 1$, $|K| + 2, \ldots$. In this expression B_x is a rotational constant and $E_x(K)$ is the lowest value for any level in the resulting "band" of energies identified by other quantum numbers x. In such systems the selection rules for electromagnetic transitions require both $|\Delta J| \leq M$ and $|\Delta K| \leq M$, where M is the multipolarity of the transition.

In most cases of interest, the lifetime of the isomeric state is large because it has a value of K differing considerably from those of lower levels to which it would, otherwise, be radiatively connected. As a consequence, bandwidth funneling processes such as shown in Fig. 1c must span substantial changes in ΔK and component transitions have been expected to have large, and hence unlikely, multipolarities.

Attempts to confirm these rather negative expectations in an actual experiment have been confounded by the rarity of the 29 candidate isomers of interest for a gamma-ray laser. Experiments[9] in which the simpler cycle of Fig. 1b was pumped through a change of $\Delta J = 4$ or 5 with a pulsed source of continua, at first confirmed these reservations, showing an integrated cross section of only 10^{-25} cm^2 eV. Such values implied that one of the constituent transitions was significantly hindered as was expected for nuclei in which K and J remain good quantum numbers at all energies of relevance. The corresponding partial width was only 37 μeV, again tending to confirm the order of magnitude for the rule-of-thumb, Eq. (1). Dogma would insist that partial widths decrease further as the values of ΔK needed for transfer would be increased.

From this perspective the candidate isomer $^{180}Ta^m$ is the one of the 29 that is the most initially unattractive as it has the largest change

of angular momentum between isomer and ground state, δK. However, because it was the only isomer for which a macroscopic sample was readily available, $^{180}Ta^m$ became the first isomeric material to be optically pumped to a fluorescent level.

This particular one of the 29 candidates for a gamma-ray laser, $^{180}Ta^m$, carries a dual distinction. It is the rarest stable isotope occurring in nature[13] and it the only naturally occurring isomer.[14] The actual ground state of ^{180}Ta is 1^+ with a half-life of 8.1 hours while the tantalum nucleus of mass 180 occurring with 0.012% natural abundance is the 9^- isomer, $^{180}Ta^m$. It has an adopted excitation energy of 75.3 keV and half-life in excess of 1.2×10^{15} years.[14] Deexcitation of the isomer is most readily affirmed by the detection of the x-rays from the $_{72}Hf$ daughter resulting from decay of the ^{180}Ta ground state with an 8.1 hour half-life.

The target used in these experiments[15] conducted at the end of 1987 was enriched to contain 1.2 mg of $^{180}Ta^m$ in 30 mg of ^{181}Ta. Deposited as a dusting of oxide near the center of the surface of a 5 cm disk of Al and overcoated with a 0.25 mm layer of Kapton, this sample was believed free from self-absorption of the x-rays from the daughter Hf.

Figure 5 shows the spectra of the enriched target before and after 4 hours' irradiation with the bremsstrahlung from a LINAC having a 6 MeV end point energy. Figure 6 shows the dependence upon time of the counting rate observed in the Hf(K_α) peaks after irradiation. Data points are plotted at the particular times at which the instantaneous counting rate equals the average counting rate measured over the finite time interval shown. The figure shows the close agreement of the measured rates to the decay expected for a half-life assumed to be 8.1 hours.

From these data and the calibrated dose from the pump shown in Fig. 7, the integrated cross section for the deexcitation of the isomer can be readily calculated if the reaction is assumed to occur through a gateway state narrow in comparison to the range of energies spanned by the irradiation. A <u>minimum value</u> of $\sigma\Gamma = 4.8 \times 10^{-22}$ cm^2 eV is obtained for the integrated cross section if the gateway energy is assumed to be near 2.0 MeV. Even larger cross sections would result from the assumption that the gateway lies at higher energies where the pumping flux is decreased. This is an enormous value exceeding anything reported for

any interband transfer by two orders of magnitude. In fact, it is 10,000 times larger than the values measured for nuclei usually studied in our work. Moreover, the relatively straightforward analysis shown schematically in Fig. 8 leads to rather astonishing conclusions.

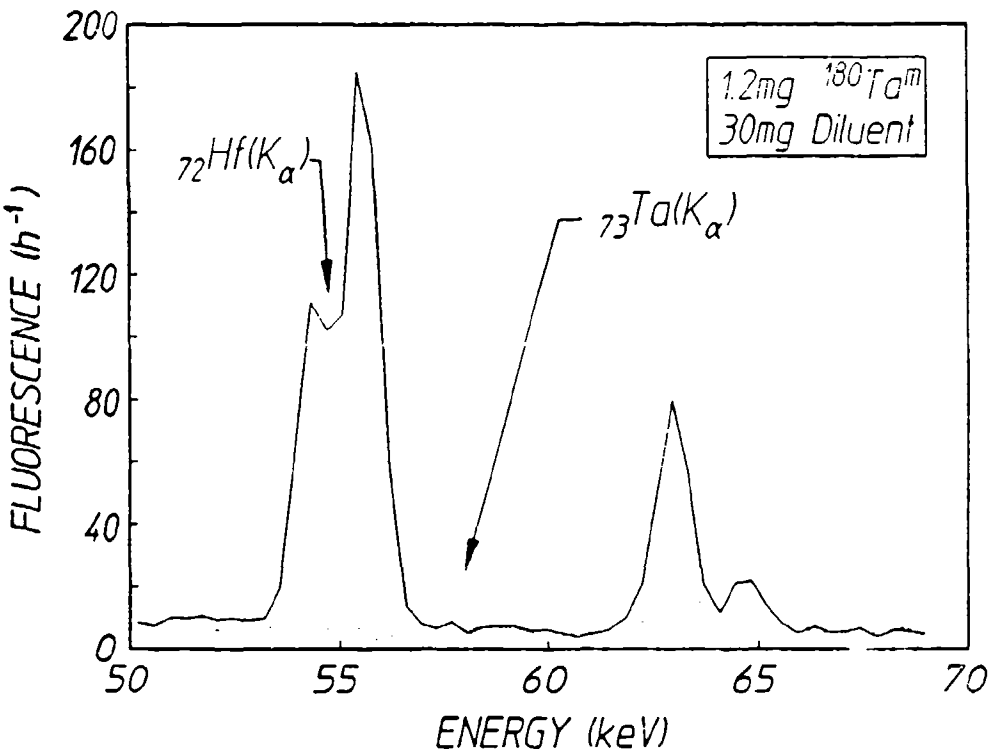

Figure 5: Dotted and solid curves show, respectively, the spectra obtained before and after dumping some of the isomeric $^{180}Ta^{m}$ contained in a target sample enriched to 5%. An HPGe detector was used to obtain the dotted spectrum before irradiation. The feature at 63 keV is from traces of natural activity in the counting shield. The solid curve shows activity resulting from the trans-mutation of the pumped ^{180}Ta measured in the same sample and counting system after irradiation. The prominent additions are the K_{α} and K_{β} hafnium x-ray lines resulting from electron capture in the ^{180}Ta.

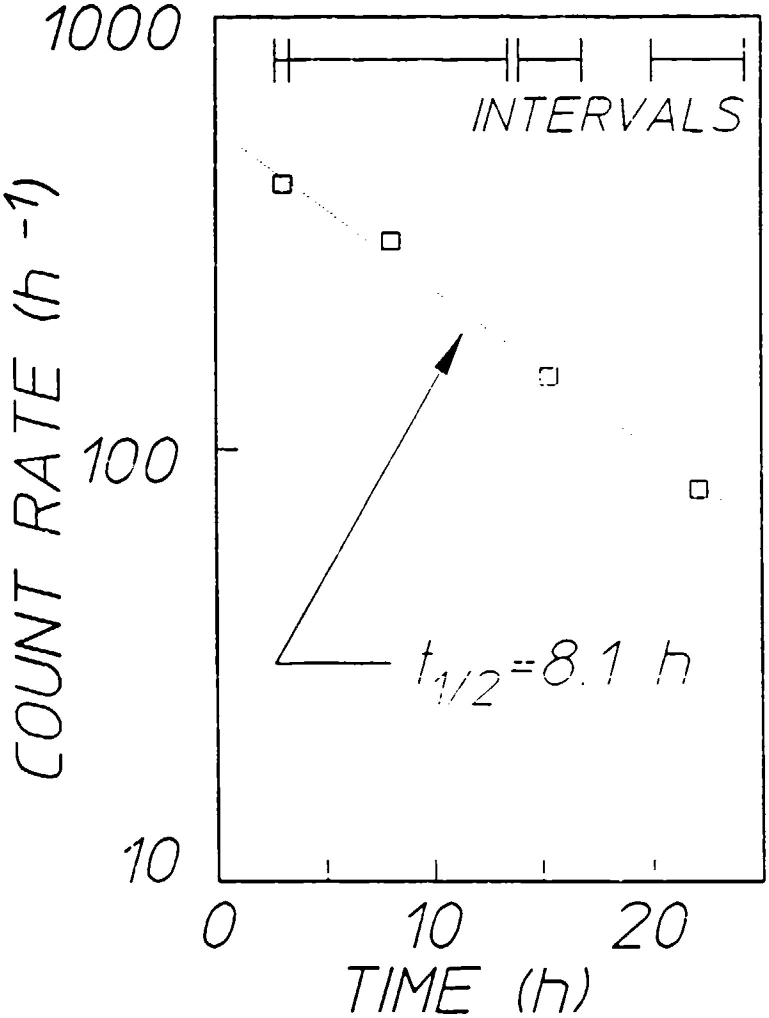

Figure 6: Plot of the counting rates measured for the Hf(K_α) fluorescence from the target as functions of the time elapsed from the end of the irradiation. The vertical dimensions of the data points are consistent with 1σ deviations of the measured number of counts accumulated during the finite counting intervals shown at the top of the graph. The dotted line shows the rate expected for a half-life of 8.1 hours.

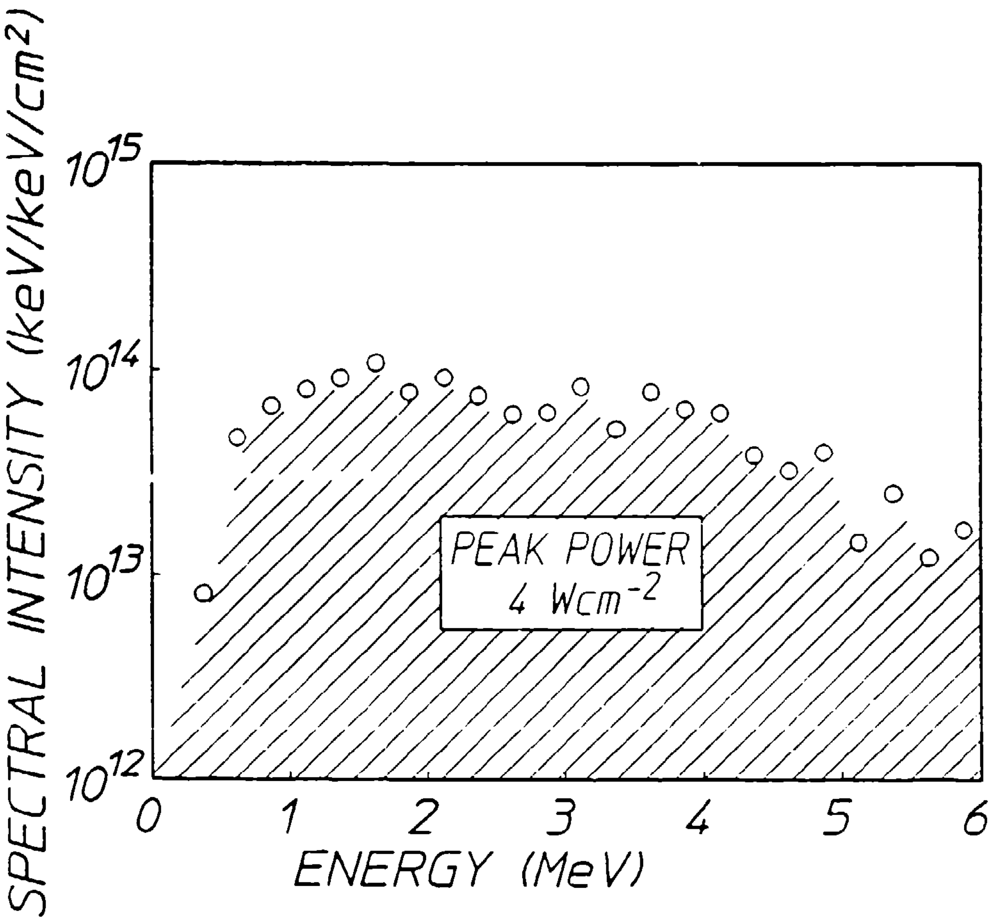

Figure 7: Spectral intensity of the bremsstrahlung used for irradiating the tantalum targets. Obtained as the total fluence from a number of successive pulses, the integral over all wavelengths of illumination corresponds to a peak power in any single pulse of only 4 W/cm^2.

Along the path of analyses of Fig. 8, assumptions are shown in ovals and derived results in rectangles. The most conservative results continue to be obtained by supposing the energy of the gateway band to which absorption first occurs to lie around 2 MeV. As shown in Fig. 8, this assumption together with the measured number of decays of ^{180}Ta gives the value being reported for the integrated cross section, $(\pi b_a b_o \Gamma \sigma_o / 2)$.

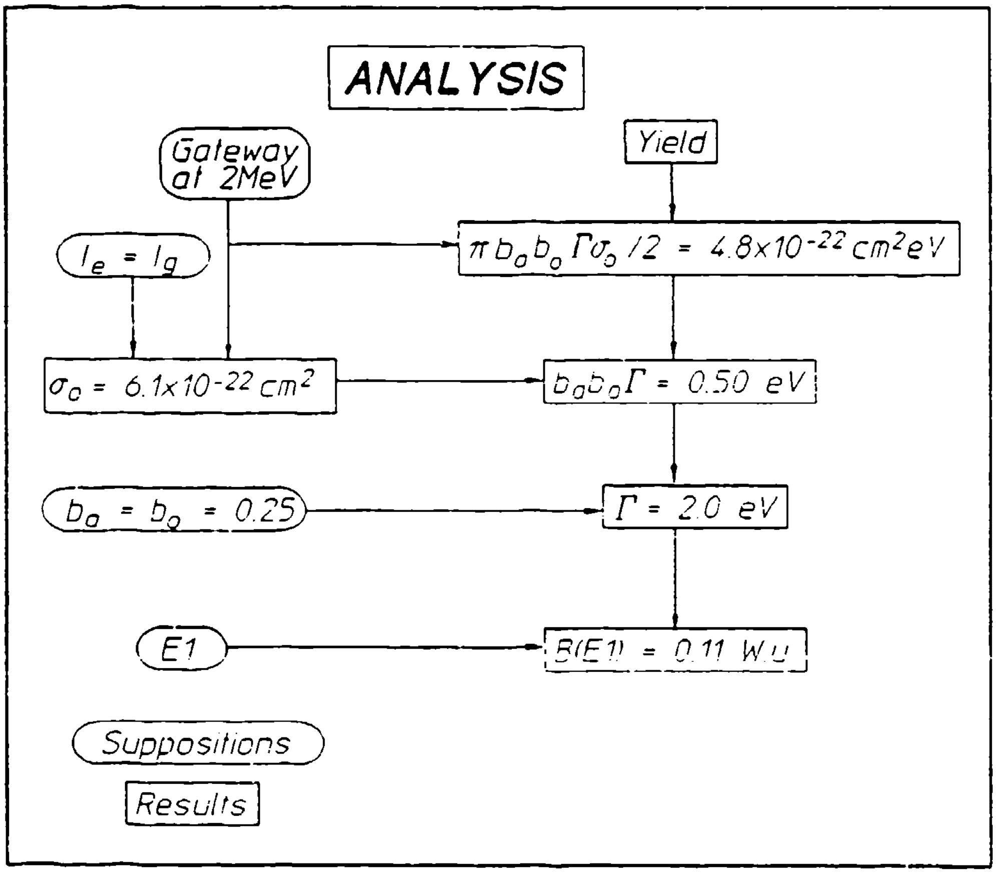

Figure 8: Flow chart showing the interrelation of assumptions and conclusions reached in the analysis of the tantalum data.

To obtain the partial width in the third row of Fig. 8 requires the Breit-Wigner cross section which peaks at

$$\sigma_o = \frac{\lambda^2}{2\pi} \frac{2I_e+1}{2I_g+1} \frac{1}{\alpha_p+1} \quad , \qquad (4)$$

where λ is the wavelength in cm of the gamma ray at the resonant energy, E_i; I_e and I_g are the nuclear spins of the excited and ground states, respectively; and α_p is the total internal conversion coefficient for the two-level system shown in Fig. 1a. The value of α_p is essentially zero for a 2 MeV transition which is highly allowed; and even were it not, σ_o would be reduced further and the partial width would become even larger. Nothing is known about the spin of the gateway state, but it is most reasonable to expect it to lie between that for the initial and

final states. In that case $I_e < I_g$, since the process is starting on the 9⁻ state. From Eq.(4) it can be seen that the assumption $I_e = I_g$ results in a probable overestimation of σ_0 and again, in an underestimation of partial width. Even underestimated in this way, the partial width for pumping the isomer down to the ground state is an astonishing,

$$b_a b_o \Gamma = 0.5 \text{ eV} \quad . \tag{5}$$

With this result of Eq.(5) the guideline of Eq.(1) is completely destroyed as a meaningful rule. *The tenet of faith limiting to 1 μeV the partial widths for pumping isomers to radiating states has been proven to be nearly a million times too pessimistic.* An extraordinary result in itself, it implies yet another unexpected feature. If analyzed further as describing the width of a single state coupled to the isomer and <u>toward</u> the ground as shown in Fig. 1c, it must be concluded that the width of the gateway state is at least 2.0 eV, as shown in Fig. 8. From the uncertainty principle,

$$\Gamma = \hbar/\tau_i \quad , \tag{6}$$

where τ_i is the lifetime of the funneling state, it is found,

$$t_{1/2}(\text{gateway}) = 0.22 \text{ fs} \quad . \tag{7}$$

To be consistent with the assumption $b_a = b_o = 0.25$ it must be concluded that the total width of 2.0 eV for the funneling level is contributed equally by two transitions, each of 1 eV width. As shown in Fig. 2, one must connect to the isomer and one to some other level with angular momentum more nearly comparable to that of the ground. Transition strengths are often measured in Weisskopf units (W.u.) since 1.0 W.u. is the maximum possible for the transition of a single nucleon for a given multipolarity.[16] Converted into those units the transition probability B(M) for one of the component steps of 1 eV width would become,

$$B(E1) = 0.038 \text{ W.u.} \quad , \tag{8a}$$

and

$$B(M1) = 6.0 \text{ W.u.} \quad , \tag{8b}$$

respectively, depending upon whether the multipolarity M were E1 or M1.

Again, these are enormous strengths, being almost without precedent. The expected[17] value for an electric dipole transition lies in the range 5×10^{-7} to 6×10^{-5} W.u. for heavy nuclei and fewer than ten are known[17,18] to approach 0.1 W.u. at these energies. For those exceptional cases, the width of the upper level is entirely due to the contribution from a single transition. Prior to the results being reported here there were no cases known where two transitions of such strength added comparable components of width to the same upper state.

The situation is little changed if the transitions are assumed to be mediated by the magnetic dipole, M1 operator. Generally, not as hindered as E1 transitions,[17] M1 strengths approach 0.1 W.u. in many cases. However, the scale of the W.u. for an M1 transition is smaller in physical units of width; so our measured widths correspond to a much larger number of W.u., thus presenting the equivalent problem. Fewer than ten M1 transitions are known[17] to have $B(M1) > 1.0$ W.u. and none are paired to share a common level.

While the width of the transfer process is difficult to interpret in the context of a single funneling state in a single particle model, a puzzle of comparable complexity is found in the efficiency with which ΔK is transferred. We have not yet been able to conceive of a cascade in the framework of pure single particle states from the funneling level to the ground state of $^{180}Ta^m$ which neither: 1) provides a "short circuit" of the flow of population between successive levels back to the initial isomeric state, nor 2) depends upon a transition away from the funneling state that would span a smaller change of energy and thus would require an even greater strength in W.u., nor 3) shortens the lifetime of the isomer by requiring the existence of a level having energy below that of the isomer and a value of J little different from 9. The width could be reduced by assuming the pumping proceeded through 1000 funneling states of comparable energy, but then the p oblem would remain that each had to support the transfer of a value of ΔJ which is difficult to accept even as a unique accident.

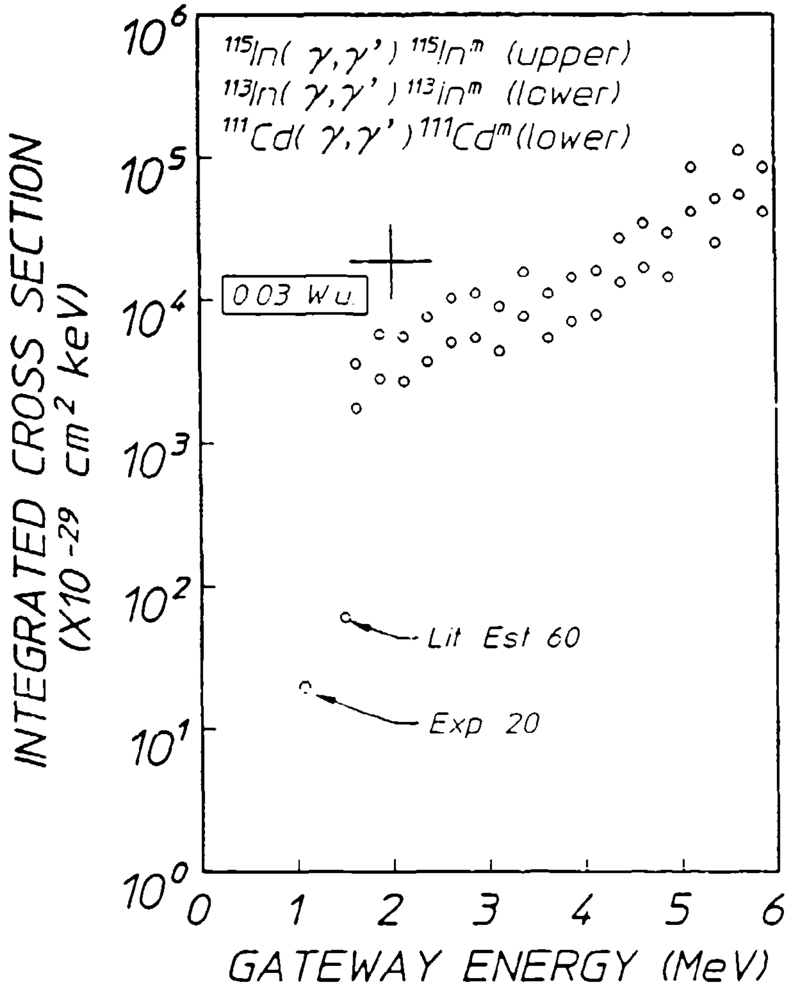

Figure 9: The integrated cross sections measured for the photoactivation of selected nuclei through individual, unknown gateway states as functions of the energies at which they could be assumed to lie. The lower family of points approximated the results obtained for the activation of both ^{111}Cd and ^{113}In to within the plotted sizes of the data. Shown at the successively lower energies are the two points taken from Refs. 11 and 7, respectively. Shown for comparison is the value corresponding to the excitation of a gateway coupled by two equal E1 transitions of the strength shown.

It is an interesting speculation that at certain energies of excitation collective oscillations of the core nucleons could break some of the symmetries upon which rest the identification of the pure single particle states. If single particle states of differing K were mixed in this way, the possibility for transferring larger amounts of ΔK with greater partial widths might be enhanced. Some support for such a speculation was found in the unexpected enhancements measured for the excitation of ^{167}Er, as discussed in the last quarterly report.[19]

If the breadth of the partial cross section for interband transfer were dependent upon a collective property, a very large integrated cross section for pumping isomers from ground state nuclei might be found to be only slightly dependent upon the detailed single particle assignments of neighboring nuclei. Just such an effect was reported[20] in our group. Integrated cross sections of the order of 10,000 in units of 10^{-29} cm^2 keV were found for the excitation of isomers of ^{111}Cd, ^{113}In, and ^{115}In through resonant gateways pumped by bremsstrahlung from a linear accelerator producing most of its intensity near 2 MeV.

Figure 9 shows the resulting values of integrated cross sections as functions of the energy E_i at which the dominant funneling state may lie. The trend in the data reflects the fact that the accelerator produced fewer photons at the higher energies, reaching zero at 6 MeV. From Fig. 9 it can be seen that integrated cross sections for the excitation of isomers of indium and cadmium reach 10^{-22} cm^2 eV for pumping through channels open to the bremsstrahlung from a 6 MeV linear accelerator. This is three orders of magnitude greater than values characteristic of excitation with photons of energy below 1.4 MeV. Shown for scale is the value of cross section which would correspond to the excitation of a gateway coupled by two equal E1 transitions of the strength shown. The similarity of results for nuclei with both similar and dissimilar single particle structures does seem to support the identification of this strong channel for optically pumping isomers with some type of core property varying only slowly among neighboring nuclei.

In a most recent effort detailed in a following manuscript, more nuclei were found to support the pumping of isomers through these enormous integrated cross sections approaching or exceeding 10^{-22} cm^2 eV. As will be seen, the partial widths nearly anticorrelate with the change in ΔJ. The largest remains ^{180}Ta^m with a change of $\Delta J = 8$, but the next

is ^{195}Pt with about a quarter of the cross section for $\Delta J = 6$. Somewhat smaller bandwidth funnels were found for nuclei for which $\Delta J = 4$.

Whatever the mechanisms, the experimental fact remains that inter-band transfer processes reaching isomeric levels can be pumped through enormous partial widths reaching 0.5 eV, even when the transfer of angular momentum must be as great as $\Delta J = 8$. Elucidation of the process, together with identification of the gateways, has been propelled into a place of importance for 1988. The most available of the isomeric candidates for a gamma-ray laser, $^{180}Ta^m$, was shown to benefit greatly from this facility for bandwidth funneling. Successfully pumped with bremsstrahlung pulses having peak intensity of only $4W/cm^2$, the great width for the transfer in $^{180}Ta^m$ provided for adequate fluorescence signals from a milligram of isomer. This fixes a pragmatic scale for the evaluation of the other 28 candidates whenever samples become available in milligram quantities.

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DEPOPULATION OF THE ISOMERIC STATE $^{180}Ta^m$ BY THE REACTION $^{180}Ta^m(\gamma,\gamma')\,^{180}Ta$

by C. B. Collins, C. D. Eberhard, J. W. Glesener, and J. A. Anderson

The isotope $^{180}Ta^m$ carries a dual distinction. It is the rarest stable isotope occurring in nature[1] and it is the only naturally occurring isomer.[2] The actual ground state of ^{180}Ta is 1^+ with a half-life of 8.1 h while the tantalum nucleus of mass 180 occurring with 0.012% abundance is the 9^- isomer, $^{180}Ta^m$. It has an adopted excitation energy of 75.3 keV and half-life in excess of 1.2×10^{15} years.[2]

The stellar s-process[3,4] for nucleosynthesis has steadily gained favor for the production of $^{180}Ta^m$ and the role of the most critical intermediary, $^{180}Hf^m$, has been well established.[2,5] However, the viability of this cosmic mechanism rests upon the absence of any reactive channel $^{180}Ta^m(\gamma,\gamma')^{180}Ta$ which could destroy the isomeric population in the photon bath present in the stellar interior at the time of creation. Prior experiments[6,7] have failed to show such a channel having any gateway for excitation below 1332 keV, but the rarity of the target material limited the sensitivity of those measurements. Reported here is the measurement of a very large cross section for the photonuclear deexcitation of $^{180}Ta^m$ through a gateway level at an energy $E \geq 1.4$ MeV. This definitive observation of such a strong radiative coupling between isomeric and ground states of ^{180}Ta may affect explanations for the natural occurrence of $^{180}Ta^m$.

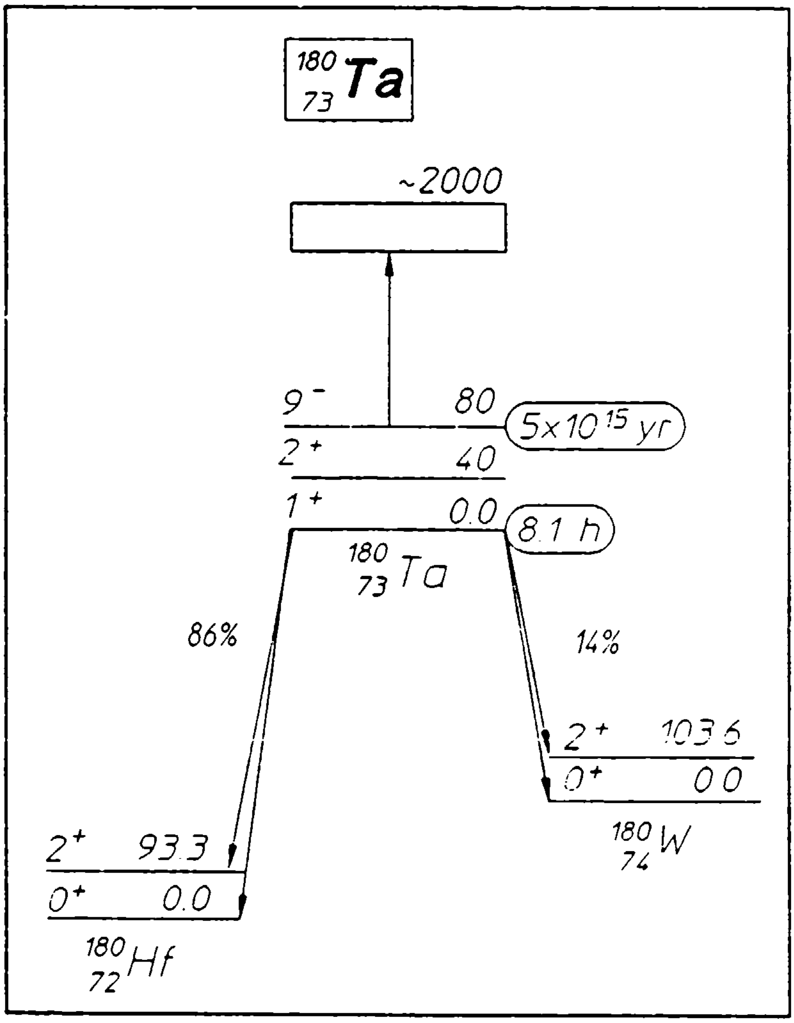

Figure 1: Schematic energy level diagram of ^{180}Ta and its daughters. Half-lives are shown in ovals to the right of the ground and isomeric levels. Energies are in keV. The initial transition of the (γ,γ') reaction is shown by the arrow pointing upward to the broad state represented by the rectangle. Cascade through the levels of ^{180}Ta is not known, but leads finally to the ground state. Electron capture to the left and beta decay to the right are indicated by the diagonal downward arrows. The final debris from pumping down the isomer is found principally in the K_α fluorescence from the ^{180}Hf characterized by the 8.1 hour lifetime of its ^{180}Ta parent.

The energy level diagram of ^{180}Ta and its daughters is shown in Fig. 1, together with a schematic representation of the individual steps in the excitation and detection of the $^{180}Ta^m(\gamma,\gamma')^{180}Ta$ reaction. As can be seen in Fig. 1, the principal means for the detection of the ^{180}Ta ground state lies in observing the K_α lines of its daughter, ^{180}Hf, following the decay by electron capture of the parent ^{180}Ta. The efficiency for the emission of K_α photons relative to the number of ^{180}Ta decays is about[8] 57%.

Two targets were used in these experiments. One consisted of a disk 5 cm in diameter of tantalum in natural isotopic abundance. It contained about 0.5 mg of $^{180}Ta^m$ in the surface layer of thickness equal to the mean distance for escape of a 55 keV x-ray photon. The second target was enriched to contain 1.3 mg of $^{180}Ta^m$ in 24.7 mg of ^{181}Ta. Deposited as a dusting of oxide near the center of the surface of a 5 cm disk of Al and overcoated with a 0.25 mm layer of Kapton, this second sample was believed[9] free from self-absorption of the x-rays from the daughter Hf.

The samples were exposed to bremsstrahlung radiation from a Varian Clinac 1800 linear accelerator (LINAC) operated with an end-point energy of 6 MeV. This device has been well characterized,[10,11] and its output dose rate has been calibrated with an accuracy of ± 3%. After irradiation, the samples were counted with an N-type, HPGe spectrometer having a beryllium entrance window. Conventional techniques were used to calibrate the counting system with isotopic standards.

Figure 2 shows the spectra of the enriched target before and after 4 hours irradiation. The spectrum from the other target was entirely similar with the Hf signal reduced by the ratio of the masses of the $^{180}Ta^m$ and the background increased by the appearance of K-lines of Ta excited in the large mass of diluent ^{181}Ta by the decay of natural activity in the counting shield.

Figure 3 shows the dependence upon time of the counting rate observed in the $Hf(K_\alpha)$ peaks after irradiation. Data points are plotted at the particular times at which the instantaneous counting rate equals the average counting rate measured over the finite time interval shown. The figure shows the close agreement of the measured rates to the decay expected for a half-life assumed to be 8.1 hours.

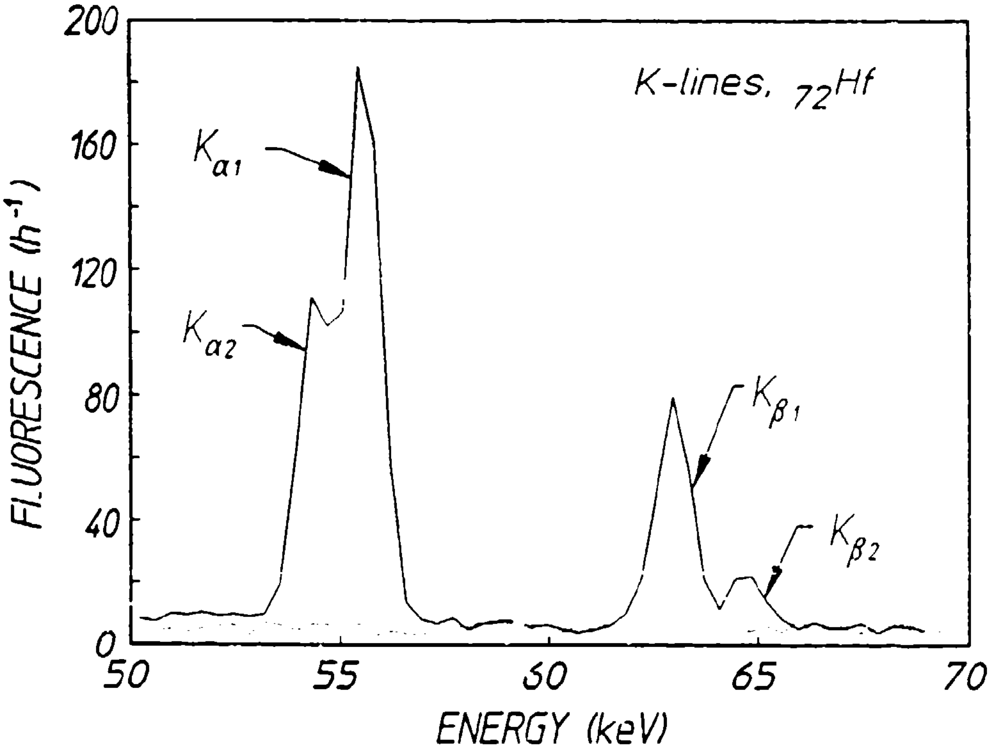

Figure 2: Dotted and solid curves show, respectively, the spectra obtained before and after dumping some of the isomeric $^{180}Ta^m$ contained in a target sample enriched to 5%. An HPGe detector was used to obtain the dotted spectrum before irradiation. The feature at 63 keV is from traces of natural activity in the counting shield. The solid curve shows activity resulting from the transmutation of the pumped ^{180}Ta measured in the same sample and counting system after irradiation. The prominent additions are the K_α and K_β hafnium x-ray lines resulting from electron capture in the ^{180}Ta.

The spectrum of the bremsstrahlung pumping the fluorescence seen in Fig. 2 was taken from the literature[10] and normalized to the total dose measured in this experiment. In this way the time integrated spectral intensity producing the fluorescence was found to be constant[12] to within a factor of two over the range 1-5 MeV at a value of 2×10^{14} keV/keV/cm^2. The number of counts observed in the Hf K_α lines were corrected for finite irradiation and counting times, the absolute counting efficiency of the spectrometer, and the 57% emission intensity from the parent ^{180}Ta to obtain the number of nuclei pumped to the ground state. Assuming self-absorption in the enriched target to be negligible, the integrated cross section for the deexcitation of the isomer can be readily calculated if the reaction is assumed to occur through a gateway state narrow in comparison to the range of energies spanned by the irradiation. A value of $\sigma\Gamma = 4.8 \times 10^{-25}$ cm^2 keV is obtained for the integrated cross section if the gateway energy is arbitrarily assumed to be near the lowest value consistent with prior[7] negative results, 2.0 MeV. Even larger cross sections would result from the assumption that the gateway lies at higher energies where the pumping flux is decreased or from from inclusion of an exact self-absorption correction. Once the gateway energy is fixed, experimental error in the integrated cross section is bounded on the lower side by a total uncertainty of 15% contributed by the calibrations of source and detector and on the upper side by a factor of two arising from the possible loss of signal because of self-absorption of the Hf x-rays.

The results of this work show a radiative connection between the isomer ^{180}Ta^m and the ^{180}Ta ground state of remarkable strength. Comparative values for the deexcitation of other isomers are not available as it appears this is the first such measurement. However, the inverse process for the excitation of isomers by (γ,γ') reactions typically proceed[13-15] with integrated cross sections at least two orders of magnitude smaller. The value reported here for the reaction ^{180}Ta$^m(\gamma,\gamma')^{180}$Ta is inexplicably large and may have several consequences. If the gateway level through which it proceeds is not sufficiently above the thermal energies expected to characterize the s-process of nucleosynthesis, current models of the stellar production of ^{180}Ta^m will be severely affected.

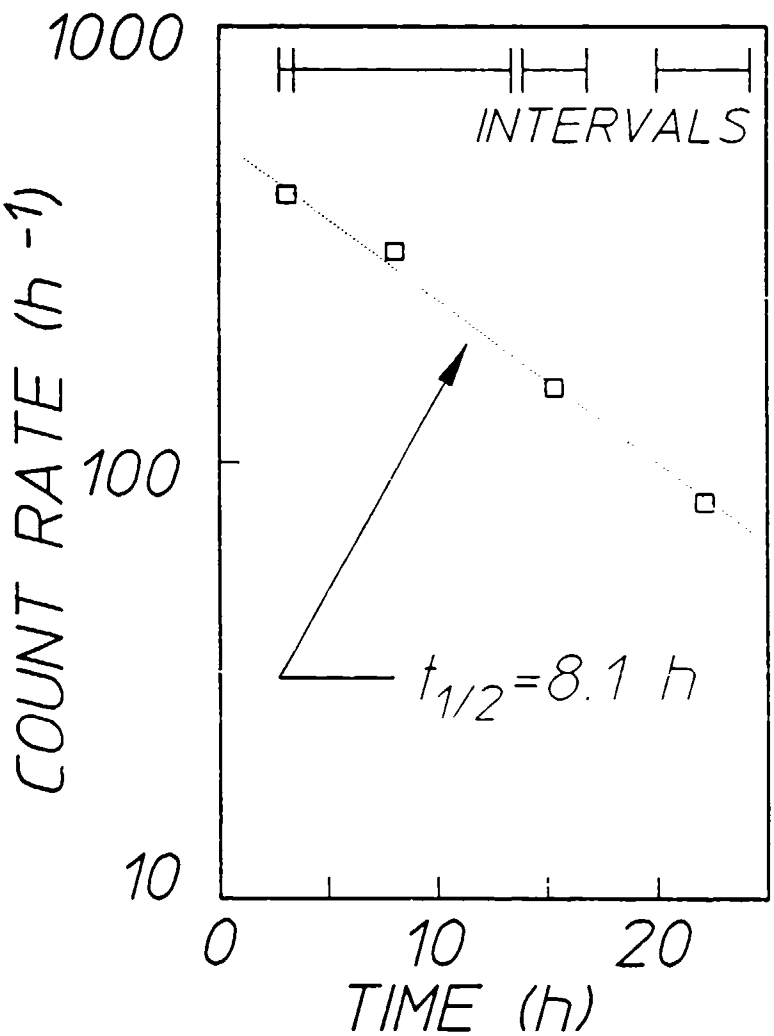

Figure 3: Plot of the counting rates measured for the Hf(K_α) fluorescence from the target fabricated from natural tantalum as functions of the time elapsed from the end of the irradiation. The vertical dimensions of the data points are consistent with 1σ deviations of the measured number of counts accumulated during the finite counting intervals shown at the top of the graph. The dotted line shows the rate expected for a half-life of 8.1 hours.

Acknowledgement

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RADIATION ENHANCED DECAY OF $^{180}Ta^m$ NUCLEI IN STELLAR INTERIORS

by J. W. Glesener, C. D. Eberhard, and C. B. Collins

Nature's rarest[1] stable isotope $^{180}Ta^m$ has an importance far exceeding its relatively small abundance. Until quite recently,[2-6] it was not clear how there could be any natural occurrence of this material. However, in 1985 the reaction necessary for the stellar s-process was demonstrated experimentally.[2,3] From that result it could be reasonably concluded[7] that the rarity of $^{180}Ta^m$ was simply a consequence of the fact that this nuclei lay aside the main path of cosmic nucleosynthesis.

The fact that $^{180}Ta^m$ is an isomer[8] presents an additional difficulty to its survival. The viability of the s-process for producing the existing population rests upon the absence of any reactive channel, $^{180}Ta^m(\gamma,\gamma')^{180}Ta$, which could destroy the isomeric population in the photon bath present in the stellar interior at the time of creation. Prior experiments[9,10] have failed to show such a channel having any gateway for excitation below 1332 keV, but the rarity of the target material limited the sensitivity of those measurements. Upper limits on cross sections were obtained, and with those constraints a detailed analysis of the effect of the stellar temperature upon the lifetime of $^{180}Ta^m$ was reported.[10] A sufficiently large region of parameter space for survival was identified to instill confidence in the current hypotheses for the natural occurrence of $^{180}Ta^m$.

The critical work[10] in establishing the survivability of the s-process $^{180}Ta^m$ product was dependent upon two assumptions, eminently reasonable at the time they were advanced. One was that all photons above a certain gateway energy could contribute to the (γ,γ') reaction through a nonresonant process of excitation established[11,12] for analogous reactions in ^{115}In and ^{111}Cd. The other was the small limit placed on the cross sections for such reactions in $^{180}Ta^m$. The principle experiment[10] had been negative, so the assumption of a large, nonresonant width to the channel forced the upper limit on cross section to a very low value.

The most recent studies of (γ,γ')reactions have made the first assumption no longer defensible. Reexaminations[13-16] of the $^{115}In(\gamma,\gamma')^{115}In^m$ and $^{111}Cd(\gamma,\gamma')^{111}Cd^m$ reactions have shown no evidence for the importance of nonresonant channels of excitation. It has been

reported[15,16] that the previous indications of such phenomena had actually been the results of a departure of the photon source from expectations. Evidently any effects of a photon bath to equilibrate isomeric and ground state populations must occur through relatively narrow gateway levels, more usefully described in terms of a product of cross section and width.[17] The second assumption has been called into question by the recent report[18] of a very large integrated cross section for the reaction $^{180}Ta^{m}(\gamma,\gamma')^{180}Ta$. Contrary to initial assumptions, there is a radiative connection of major strength between isomer and ground, but the gateway energy is still unknown.

For deexcitation of the $^{180}Ta^{m}$ isomer through a gateway state narrow in comparison to any structure in the spectrum of the photon flux Φ, the rate is proportional to the quantity $(\sigma\Gamma)\Phi$, where the product $(\sigma\Gamma)$ is the integrated cross section for the reaction. From the yield of daughter products reported earlier[18] and knowledge[19] of the spectrum of irradiation, Φ, the possible values of $(\sigma\Gamma)$ can be obtained as functions of the value assumed for the unknown energy of the dominant gateway. The resulting possibilities of integrated cross section are shown in Fig. 1. Since the threshold energy of the nonresonant gateway was a free parameter in the original calculation[10] of the lifetime of the isomer in the stellar environment, the data of Fig. 1 serve as the needed input for a recomputation of those lifetimes for the more realistic case of a resonant gateway state through which thermal equilibrium must proceed. Those calculations are reported here.

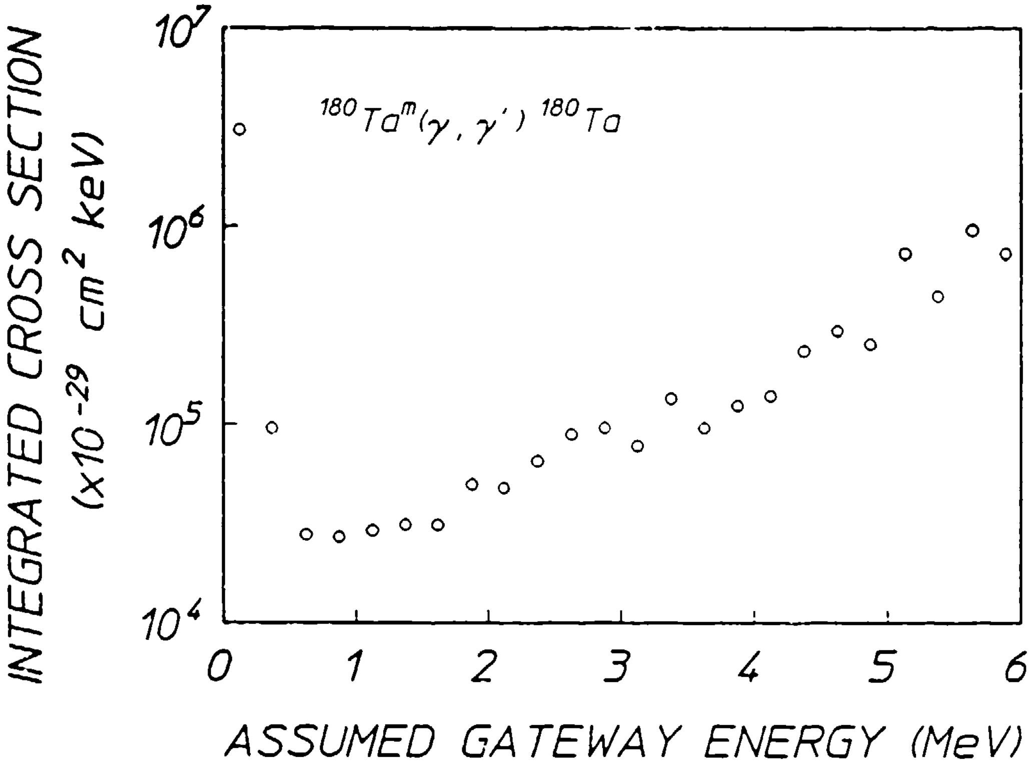

Figure 1: Plot of the integrated cross section measured for the reaction $^{180}Ta^m(\gamma,\gamma')^{180}Ta$ as a function of the energy at which the gateway state could be assumed to lie.

This isotope of tantalum can be treated as a two-level system in a radiation field.[20] The rate equations for the populations of isomeric and ground states are, respectively,

$$\frac{dN_1}{dt} = -(\lambda_1 + \lambda_{10})\,N_1 + \lambda_{01}N_0 \quad , \qquad (1a)$$

and

$$\frac{dN_0}{dt} = -(\lambda_0 + \lambda_{01})\,N_0 + \lambda_{10}N_1 \quad , \qquad (1b)$$

where

λ_1 = isomeric decay rate,

λ_{10} = transition rate, isomeric state to ground state,

λ_{01} = transition rate, ground state to isomeric state,

λ_0 = ground state decay rate,

1 = isomer state variable, and

0 = ground state variable.

In thermal equilibrium,

$$\frac{N_1}{N_o} = \frac{2J_1+1}{2J_0+1} e^{-E_1/kT} \simeq \frac{\lambda_{01}}{\lambda_{10}} \quad . \qquad (2)$$

Solving the two rate equations for N_1/N_0 yields

$$\frac{N_1}{N_0} = \frac{\lambda_c + \lambda_{01} + u \times \coth(ut) - b/2}{\lambda_{10}} \quad , \qquad (3)$$

where

$$b = (\lambda_0 + \lambda_1 + \lambda_{01} + \lambda_{10}),$$

$$c = [(\lambda_1 + \lambda_{10})(\lambda_0 + \lambda_{01}) - \lambda_{10}\,\lambda_{01}], \text{ and}$$

$$u = \frac{(b^2 - 4c)^{1/2}}{2} \quad .$$

Assuming a black body distribution in the stellar interior,

$$\lambda_{10} = \sigma\Gamma c \frac{E^2}{\pi^2(hc)^3} e^{-E/kT} \quad , \qquad (4)$$

where

$\sigma\Gamma$ = the cross section width in cm^2-keV, and

E = the energy of the intermediate gateway state.

From Ref. 9, the definition of the effective half-life of $^{180}Ta^m$ is

$$t_{1/2}^{eff} = \left(1 + \frac{N_1}{N_0}\,(t_{1/2}^{(0)})\right) t_{1/2}^{(0)} \quad , \qquad (5)$$

where

$t_{1/2}^{(0)}$ = the lifetime of the ground state.[21]

A simple formula for $t_{1/2}^{(0)}$ is,[22,23]

$$t_{1/2}^{(0)} = \frac{1}{\dfrac{b_{ec}}{t_{1/2}} \dfrac{1}{e^{-(E/kT-\mu)} + 1} + \dfrac{b_\beta}{t_{1/2}}} , \qquad (6)$$

where

E = K shell energy,

$t_{1/2}$ = $^{180}Ta^m$ ground state half-life,

μ = chemical potential,

b_{ec} = branching ratio for electron capture, and

b_β = branching ratio for beta decay.

The resulting half-life in the stellar environment, $t_{1/2}^{eff}$, is plotted in Fig. 2 as a function of temperature for different energies which could be assumed for the gateway state. The conditions for survivability can be readily appraised. At the canonical temperature[10] of 3.5×10^8 K for the s-process, the critical question is whether the gateway for deexcitation of $^{180}Ta^m$ lies within the interval 1.4 - 2.0 MeV. If it does, then the extant population of $^{180}Ta^m$ could not have survived the temperature of creation. Conversely, location of the gateway above 2 MeV would affirm the viability of an s-process mechanism for the production of $^{180}Ta^m$.

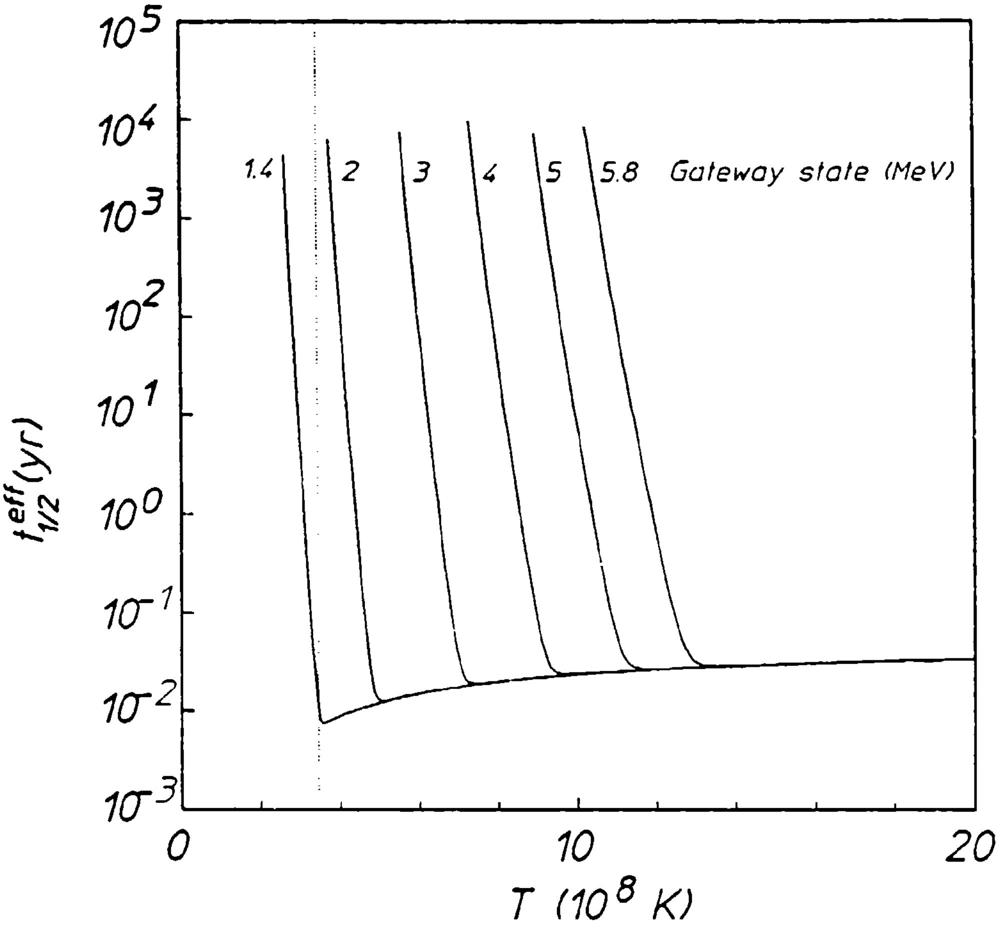

Figure 2: Graph of the effective half-life of a $^{180}Ta^m$ nucleus as a function of the temperature of the radiation bath in which it is immersed. Different curves show the results of assuming excitation and deexcitation occur through a dominant gateway state lying at the energies shown. The dotted line indicates the nominal temperature assumed to characterize the stellar s-process.

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LARGE CHANGES OF ANGULAR MOMENTA PUMPED BY BREMSSTRAHLUNG IN SELECTED NUCLEI

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A renaissance in the study of (γ,γ') reactions has been launched by the availability of medical linear accelerators (linacs) which can serve as intense and stable bremsstrahlung sources with particularly well-characterized spectra.[1,2] The total doses which they can deposit in reasonable working periods have made possible the examination of (γ,γ') reactions involving rare materials for which target masses are limited to milligrams. In this way, the first (γ,γ') reaction leading to the deexcitation of an isomeric sample was studied,[3] with rather unexpected results. Requiring an unlikely change of $\Delta J = 8$, this isomer $^{180}Ta^m$ was dumped through a partial width of at least 0.5 eV, an enormous value exceeding anything previously reported for (γ,γ') reactions at comparable energies by two to three orders of magnitude. Subsequently, the reactions producing some isomers were found[4] to proceed through nearly comparable partial widths of 0.05 eV.

The integrated cross sections for either pumping or dumping of nuclear isomers are usually expressed as $\pi b_a b_o \sigma_o \Gamma/2$ where Γ is the natural width in keV of the i-th pump band,

$$\Gamma = \hbar/\tau_p \quad , \qquad (1)$$

where τ_p is the natural lifetime and the branching ratios b_a and b_o give the probabilities for the decay of the broad level back into the initial and fluorescence level, respectively. The pump energy E_i is in keV and σ_o is the amplitude of the Breit-Wigner cross section for the absorption transition,

$$\sigma_o = \frac{\lambda^2}{2\pi} \frac{2I_e+1}{2I_g+1} \frac{1}{\alpha_p+1} \quad , \tag{2}$$

where λ is the wavelength in cm of the gamma ray at the resonant energy E_i; I_e and I_g are the nuclear spins of the excited and ground states, respectively; and α_p is the internal conversion coefficient of the absorption transition. If there is more than one pump band linking initial and final states, products of integrated cross section and input flux must be summed over the appropriate bands.

The combination, Γ_x

$$\Gamma_x = b_a b_o \Gamma \quad , \tag{3}$$

is the partial width for excitation (or deexcitation) of an isomeric sample. Because of the possibility of cascading transitions being involved in the reaction, a partial width measured in one sense does not necessarily characterize the inverse process.

Cross sections for the archetype cases[5,6] of ^{115}In and ^{111}Cd were of the order of 10 in the conventional units of 10^{-29} cm^2 keV. Such values for excitation through a gateway near 1 MeV are characteristic of products of branching ratios $b_a b_o$ somewhat degraded from the optimal value of 0.25. One transition is primarily responsible for the favorable width of the gateway. The other transition is parasitic, contributing lesser additional width. Whether this is simply coincidental for these two cases or the result of a general principle is not known.

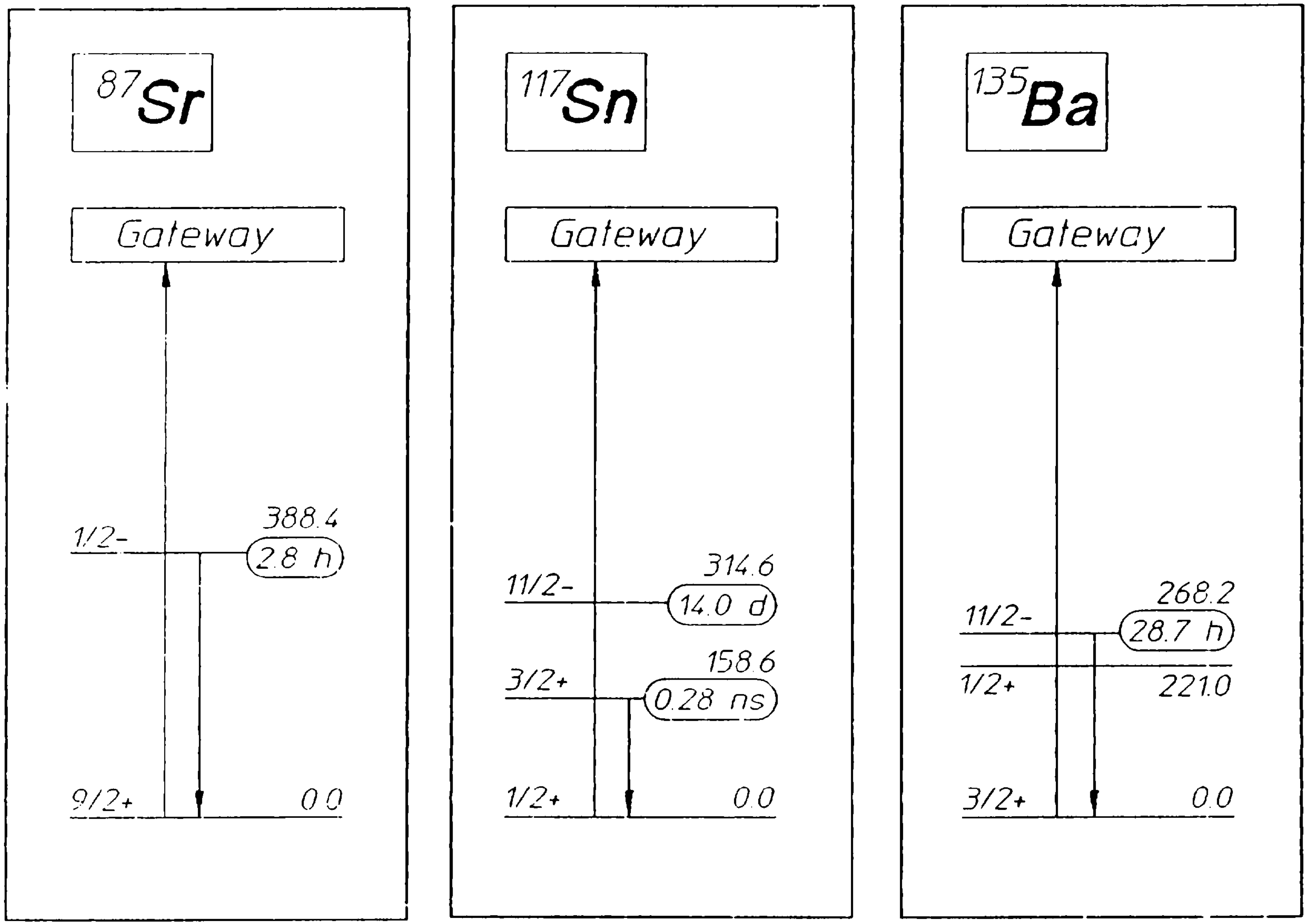
Very early data[7,8] indicated that yields from (γ,γ') reactions increased as higher energy gateway states were accessed. Evidence was accumulated[7,8] in the form of increases in the slopes of curves showing product yields as functions of the end point energies of the bremsstrahlung used to pump the reactions, but the changes were not dramatic. The largest value found[8] was 580 units for the (γ,γ') excitation of ^{87}Sr^m from the ground state through a gateway at 2.66 MeV.

Systematic studies[9] have shown that collective octupole oscillations of the nuclear core can unhinder E1 transitions, making very short lived states available for (γ,γ') reactions excited from ground states at energies between 1 and 2 MeV. However, the literature[10] suggests that the branching for such a collective state would almost entirely favor the initial transition so that a diminishing product, $b_a b_o$, would

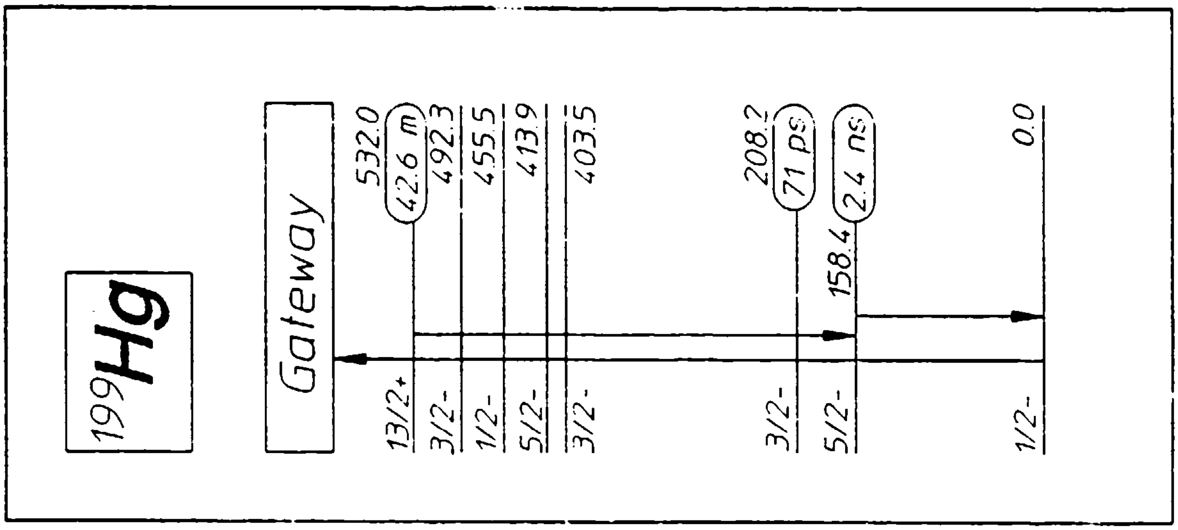
largely offset the greatly increased width Γ in expressions for the integrated cross section for a (γ,γ') reaction excited through such a collective state. Such an expectation is supported by the early data mentioned above.

Since the density of states is considerably elevated at energies of 1 to 2 MeV above the ground state, an alternate speculation is attractive. A strong collective oscillation of the core might serve to mix enough single particle states so that radiative branches to several different lower levels become comparable. In this case a very large integrated cross section of (γ,γ') reactions producing isomers from ground state nuclei might be found to be only slightly dependent upon the detailed single particle assignments of neighboring nuclei. Just such an observation was recently reported.[4] Integrated cross sections of the order of 10,000 in the units of 10^{-29} cm^2 keV were found for the excitation of isomers of ^{111}Cd, ^{113}In, and ^{115}In through resonant gateways pumped by bremsstrahlung from a linear accelerator producing most of its intensity near 2 MeV.

Reported here is the extension of these studies to the excitation of very long-lived isomers having half-lives varying from hours to weeks. Pumped with a linac having its end point energy at 6 MeV, the (γ,γ') reactions had to proceed through channels providing for changes of angular momentum ranging from $\Delta J = 4$ to 6. Five nuclides were examined: ^{87}Sr, ^{117}Sn, ^{135}Ba, ^{195}Pt, and ^{199}Hg. Integrated cross sections were found to range from 1,000 to 20,000 in the usual units of 10^{-29} cm^2 keV, the facility for excitation showing no correlation with ΔJ. The largest occurred for $^{195}Pt(\gamma,\gamma')^{195}Pt^m$ which requires $\Delta J = 6$ to excite a 4 day isomer. While the energies of the responsible gateways cannot yet be determined, the pervasiveness of such large partial widths for the exchange of substantial amounts of angular momentum was unexpected at any energies below the thresholds for (γ,n) reactions.



87Sr
Gateway
1/2-
388.4
2.8 h
9/2+
0.0
117Sn
Gateway
11/2-
314.6
14.0 d
3/2+
158.6
0.28 ns
1/2+
0.0
135Ba
Gateway
11/2-
268.2
28.7 h
1/2+
221.0
3/2+
0.0



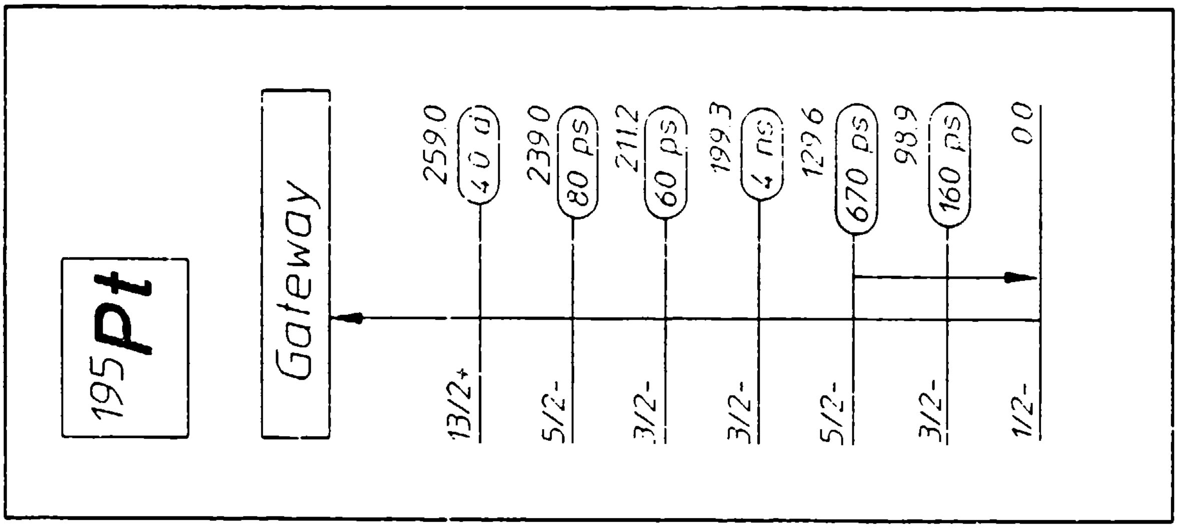


Figure 1: Energy level diagrams of the excited states important to the production and detection of the isomers of the nuclei shown. Half-lives of the states are shown to the right of each, together with their energies in keV. Downward arrows locate transitions used in the detection of populations of the isomers produced by the absorption transitions indicated by the upward arrows. The locations of the gateways through which the (γ,γ') reactions proceed are not to scale, and the details of the cascades downward to the isomers are unknown.
a), b) and c) (opposite from left): Transitions important to the study of (γ,γ') reactions producing the isomers $^{87}Sr^m$, $^{117}Sn^m$, and $^{135}Ba^m$.
d) and e) (from left): Transitions important to the study of (γ,γ') reactions producing the isomers of $^{195}Pt^m$ and $^{199}Hg^m$. For the latter, two possible transitions are shown for the detection of the isomer.

Experimental Procedures

The relevant energy levels for the five nuclei of interest in these experiments are shown in Fig. 1. They were present in targets fabricated from materials containing natural isotopic abundances. In some cases they were thick enough that self-absorption of the output transition necessitated a correction of significant magnitude. The samples of elemental Pt and Sr were in plate form. The other samples were powders held in flat, cylindrical polyethylene vials. All samples were counted at the endcap window of a 10% relative efficiency, n-type Ge detector. Target parameters are summarized in Table I, together with the corrections for self-absorption computed from the counting geometry, sample composition, and sample density. As shown in Fig. 1, two fluorescent transitions occurred with sufficient probabilities from the $^{199}Hg^m$ state to support measurement of the integrated cross section for the $^{199}Hg(\gamma,\gamma')^{199}Hg^m$ reaction. Having different energies, they encountered different levels of self-absorption and served to confirm the procedures for correction.

Table I

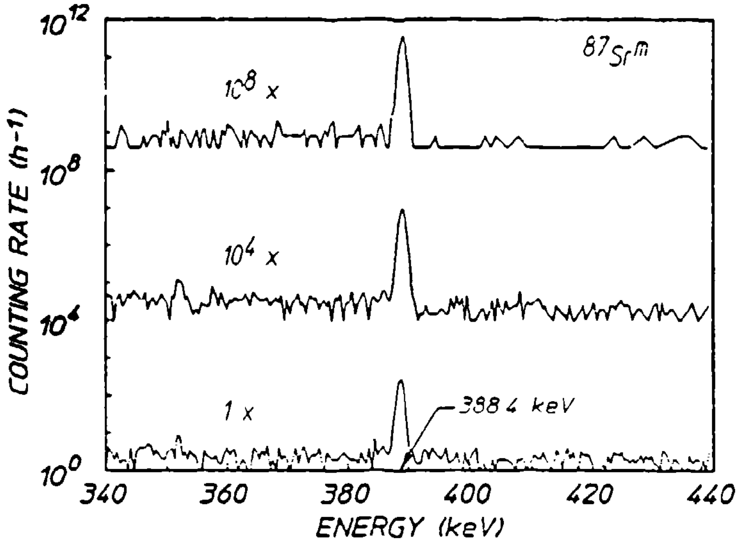
Summary of target parameters for the nuclides used in these experiments. The transparency factor listed in the rightmost column is the computed fraction of the emitted counts able to reach the spectrometer in the particular geometry employed.

Nuclides	Form	Abundance (%)	Sample Mass (g)	Half-life (days)	Fluorescence (keV)	Transparency
^{87}Sr	SrF_2	7.0	13.4	0.117	388.4	0.934
^{117}Sn	Sn	7.7	4.35	13.6	158.6	0 911
^{135}Ba	BaF_2	6.6	8.65	1.20	268.3	0.94
^{195}Pt	Pt	33.8	31.1	4.02	129.8	0.06
^{199}Hg	Hg_2Cl_2	16.9	24.0	0.030	374.1 158.5	0.337 0.41

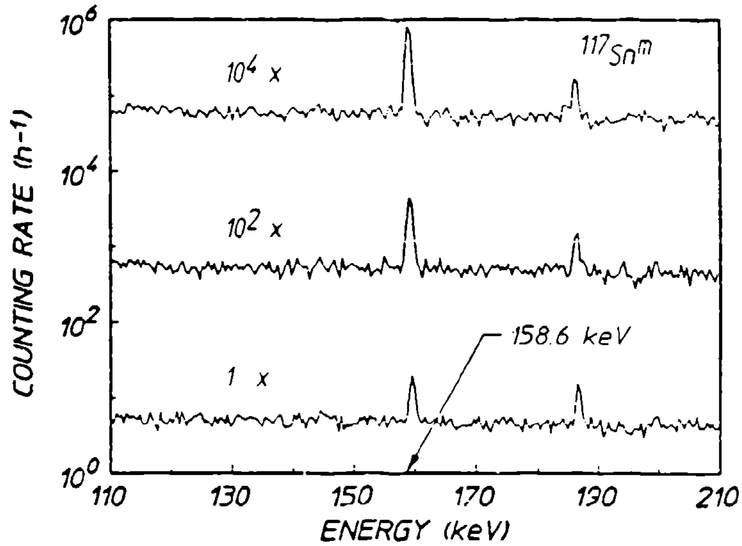
Targets were exposed for times on the order of four hours to the output of a Varian Clinac 1800 linear accelerator at the Department of Radiology of the University of Texas Southwestern Medical Center at Dallas. This linear accelerator was operated with an end point energy of 6 MeV.

After irradiation, targets were removed to the counting facility of the Center for Quantum Electronics of the University of Texas at Dallas, where the decays of the isomeric products of the (γ,γ') reactions were measured with the Ge spectrometer system. Typical spectra are shown in Figs. 2a - 2e.

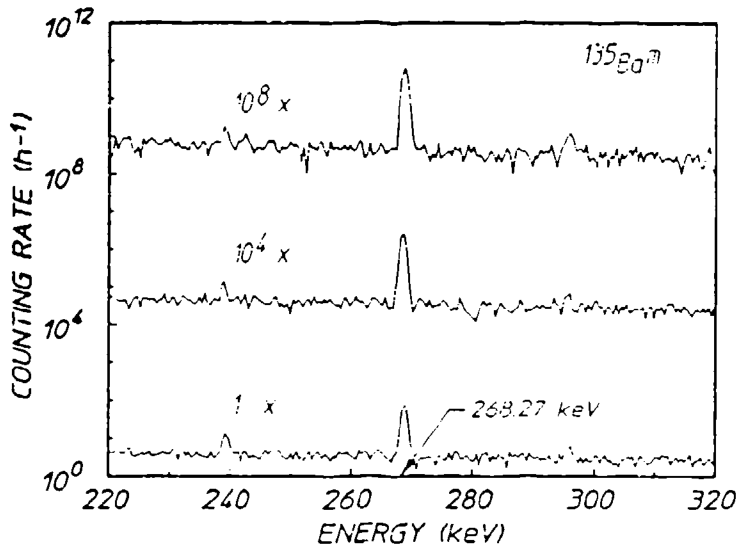
Confirmation that the fluorescence peaks were the signatures of the decays of the respective isomers was obtained by examining the decays of the counting rates as functions of the time elapsed from the cessation of the irradiation. These decay curves are shown in Fig. 3a - 3e, together with lines recording the expectations based upon literature values of the half-lives. Fluctuations expected at the 1σ level correspond roughly to the sizes of the plotted points with one exception in which the 1σ error bars are explicitly shown. For each of the five nuclides studied, both spectral and temporal content of the fluorescence conformed to the unequivocal signatures of the five isomers expected.



$^{87}Sr^{m}$
10^8 x
10^4 x
1 x
388.4 keV
10^{12}
10^{8}
10^{4}
10^{0}
COUNTING RATE (h-1)
340 360 380 400 420 440
ENERGY (keV)



$^{117}Sn^{m}$
10^4 x
10^2 x
1 x
158.6 keV
10^{6}
10^{4}
10^{2}
10^{0}
COUNTING RATE (h-1)
110 130 150 170 190 210
ENERGY (keV)



$^{135}Ba^{m}$
10^8 x
10^4 x
1 x
268.27 keV
10^{12}
10^{8}
10^{4}
10^{0}
COUNTING RATE (h-1)
220 240 260 280 300 320
ENERGY (keV)

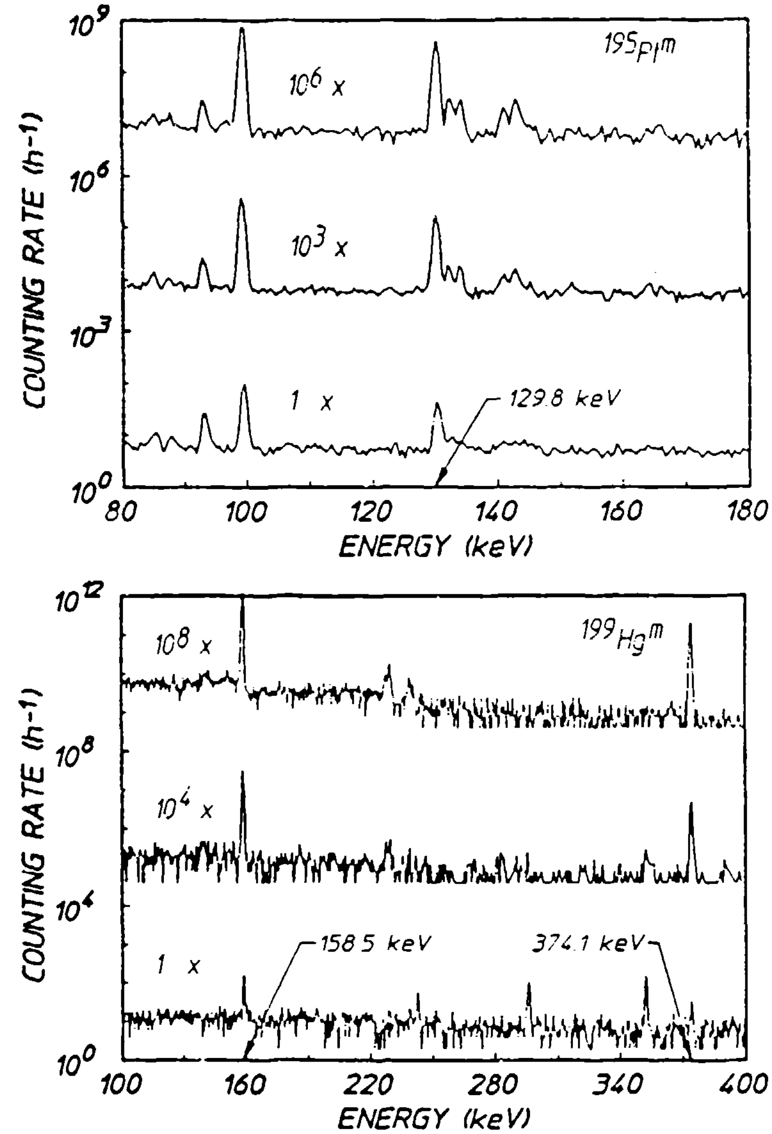


Figure 2: Successive spectra of the fluorescences detected with an intrinsic Ge detector emphasizing the ranges of energies shown. Transitions shown in Figs. 1a - 1e for the detection of the isomers are identified by the arrows. In each figure successive spectra have been offset vertically by the amounts shown, with the one taken directly after irradiation at the top. The times elapsed from the ends of irradiation to the start of the counting intervals are as follows:

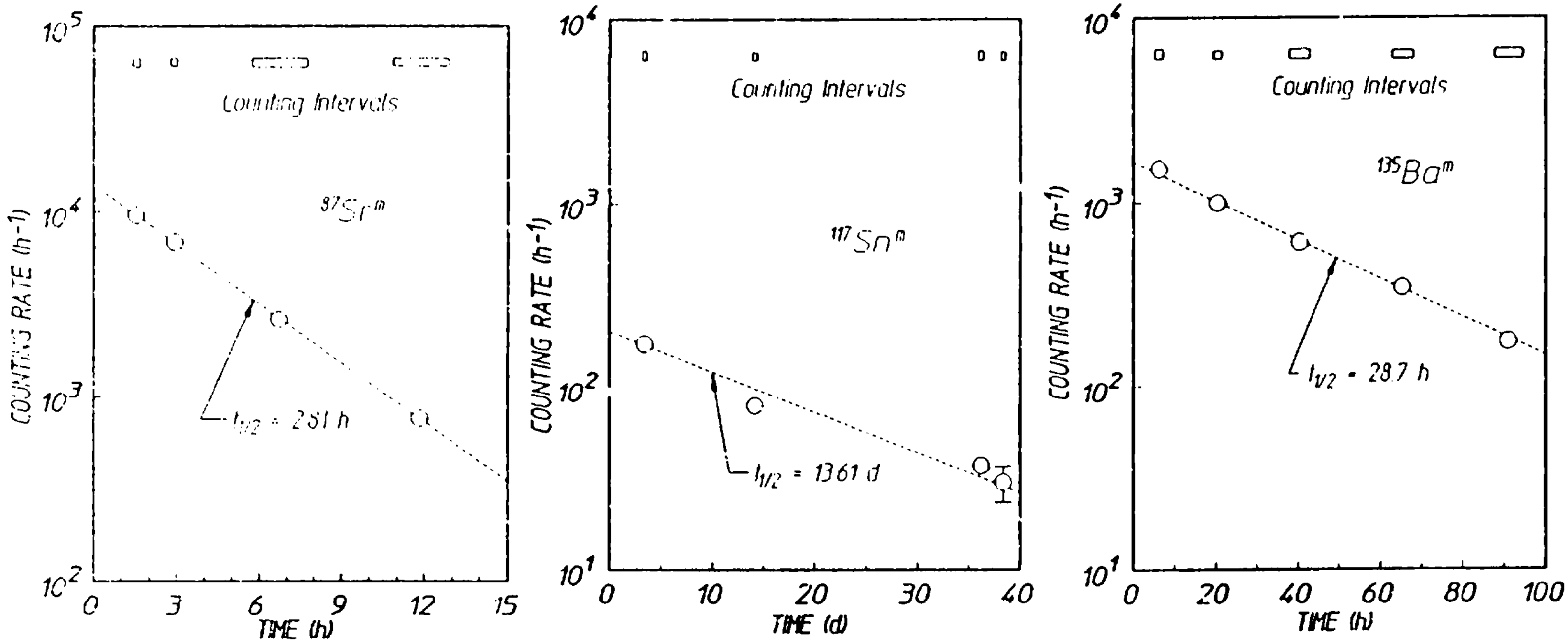
a) (opposite top) ^{87}Sr; 1.42, 5.77, and 10.87 h for durations of 15, 120, and 120 min, respectively.

b) (opposite center) ^{117}Sn; 3.17, 13.96, and 38.1 days for durations of 600 min each. The peak at higher energies is from the natural background.

c) (opposite bottom) ^{135}Ba; 5.20, 37.8, and 87.6 h for durations of 120, 300, and 402.8 min, respectively. The spurious peak at lower energies is from the natural background.

d) (top) ^{195}Pt; 0.59, 4.99, and 12.96 days for durations of 300, 600, and 600 min, respectively. The transition at 98.9 keV is also part of the cascade from the isomer $^{195}Pt^m$, originating on the lowest excited state in Fig. 1d. Structures at even lower energies are part of the background.

e) (center) ^{199}Hg; 49.97, 135.1, and 315.1 min for durations of 15, 15, and 30 min, respectively.


Counting Intervals
$^{87}Sr^m$
$t_{1/2}$ = 2.81 h
COUNTING RATE (h-1)
TIME (h)
Counting Intervals
$^{117}Sn^m$
$t_{1/2}$ = 13.61 d
COUNTING RATE (h-1)
TIME (d)
Counting Intervals
$^{135}Ba^m$
$t_{1/2}$ = 28.7 h
COUNTING RATE (h-1)
TIME (h)

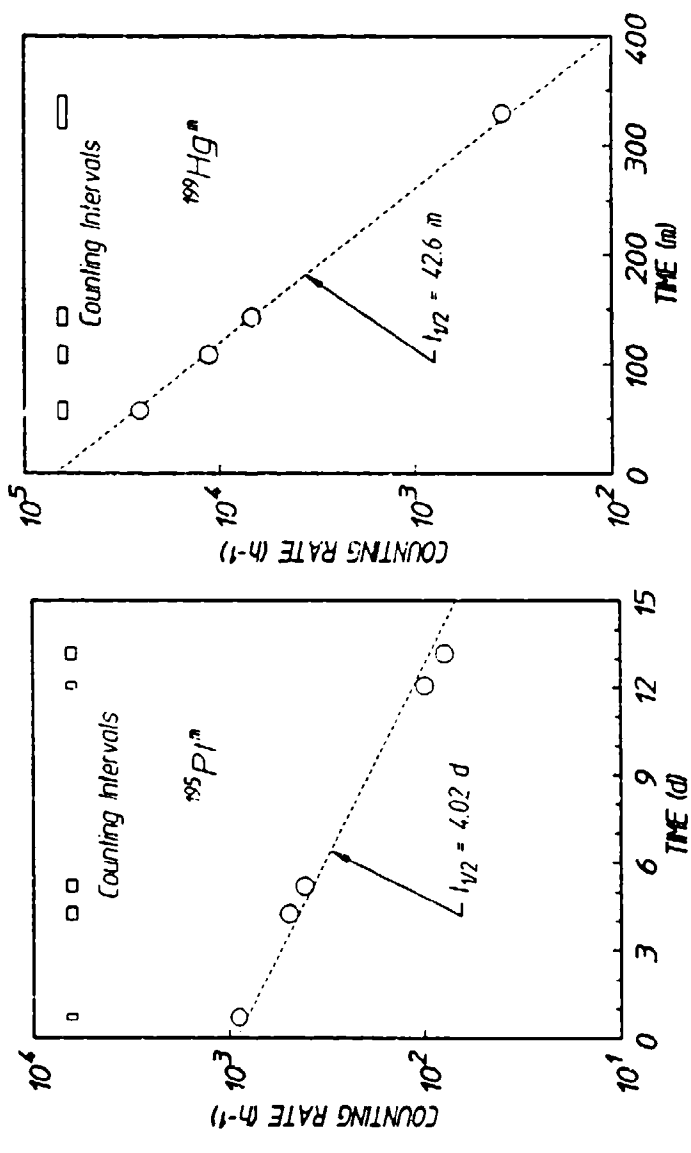


Figure 3: Plots of the counting rates measured in the peaks of the lines identified in Figs. 2a - 2e as functions of time elapsed from the end of the irradiation. Except where error bars are explicitly shown, the vertical dimensions of the data points are consistent with 1σ deviation of the measured numbers of counts accumulated during the finite counting intervals shown at the tops of the graphs. The dotted lines show the rates expected for the literature values of the half-lives.

a), b), and c) (opposite, top to bottom) Decays of the fluorescences from $^{87}Sr^m$, $^{117}Sn^m$, and $^{135}Ba^m$.

d) and e) (top and center) Decays of the fluorescences from $^{195}Pt^m$ and $^{199}Hg^m$.

Results

From the numbers of counts in the fluorescence spectra of Figs. 2a - 2e, the numbers of activations in the samples were obtained by well-established procedures. The efficiency of the spectrometer was determined with calibrated sources and was found to conform closely to nominal specifications. Self-absorption corrections were taken from Table I, and fluorescence efficiencies from the literature.[11] At this stage of analysis, the numbers of activations in ^{199}Hg indicated by the two different transitions agreed to within 2%, thus verifying the procedure for calculating the self-absorption corrections.

The rates of activations of the samples, dN/dt, were obtained by dividing the observed numbers of activations by the irradiation times after correcting for finite counting and irradiation times. Literature values of the half-lives were used in making these corrections.[12]

The excitation rate is

$$\frac{dN}{dt} = N_o \sum_i (\pi \Gamma_x \sigma_o / 2)_i \, \phi_i \qquad , \qquad (4)$$

where the parameters in parentheses comprise the integrated cross section for pumping the (γ,γ') reaction through the i-th gateway, ϕ_i is the photon flux at the energy needed to excite that gateway, and N_o is the number of target nuclei. In our experiment, the spectrum of irradiation, $\phi_i(E)$, cannot yet be varied in a controlled manner so that the sum of Eq. (4) cannot be decomposed from the experimental measurements of dN/dt into components from each of the contributing bands. An interesting alternative is to extract the effective cross section $\sigma(E)$ which would be necessary to produce the observed activation through a single gateway,

$$\sigma(E) = \frac{dN/dt}{N_o \phi(E)} \qquad . \qquad (5)$$

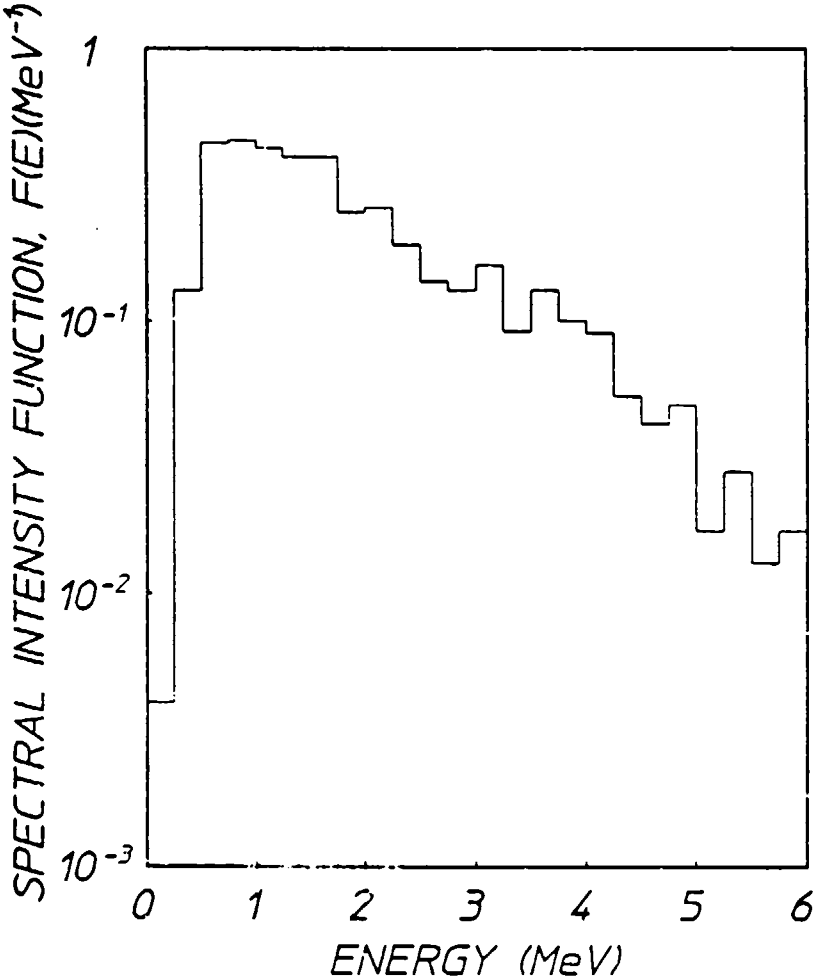


Figure 4: Relative spectral intensities of the bremsstrahlung used for irradiation in these experiments normalized so that the integral under the curve is unity.

The spectral distribution from the Clinac 1800 medical linac is considered to be well-known[1,2] and is normalized by a measurement of the total dose delivered during the irradiation. The expected distribution is shown in Fig. 4. From the $\phi(E)$ determined in this way, the effective, integrated cross section can be determined from the measured activation rate as shown in Eq. (5). The $\sigma(E)$ is a function of the energy E at which the effective gateway is assumed to lie. Results for the five isotopes examined in these experiments are shown in Figs. 5a - 5e. In each case the possible values of energies for the gateways are limited at the lower end by prior reports of much smaller cross sections for (γ,γ') reactions known to occur through gateways lying between 1 and 1.5 MeV.

Conclusions

From Figs. 5a - 5e it can be seen that the integrated cross sections for the excitations of isomers proceeding through channels open to the bremsstrahlung from a 6 MeV medical linear accelerator for these five species reach values exceeding almost all previous results by two to three orders of magnitude. However, most earlier work was conducted with sources having end point energies lying below 3 MeV so it might be initially supposed that these larger cross sections describe channels open near the threshold for (γ,n) reactions where state density is high. However, a troubling aspect is the large change of angular momentum spanned by these reactions and the lack of correlation of reaction probabilities with minimal changes in J.

The reproducibility of the experiment is demonstrated in Fig. 6, which reports the absolute level of agreement between experimental series conducted three months apart after complete disassembly and reintegration of the apparatus. Figure 7 presents a summary of the results of this work which suggests some groupings of the magnitudes of these cross sections. In the lowest group, values are reasonably continuous with prior work.[8]

The most indicative point of comparison is found in Fig. 5a reporting the cross sections for the reaction $^{87}Sr(\gamma,\gamma')^{87}Sr^{m}$. There is plo[illegible]d the largest value previously reported[8] for such a reaction, that of 280 - 580 ($\times 10^{-29}$ cm^2 keV) for the gateway at 2.66 MeV. The discrepancy between that value and our work is not greater than what was usually

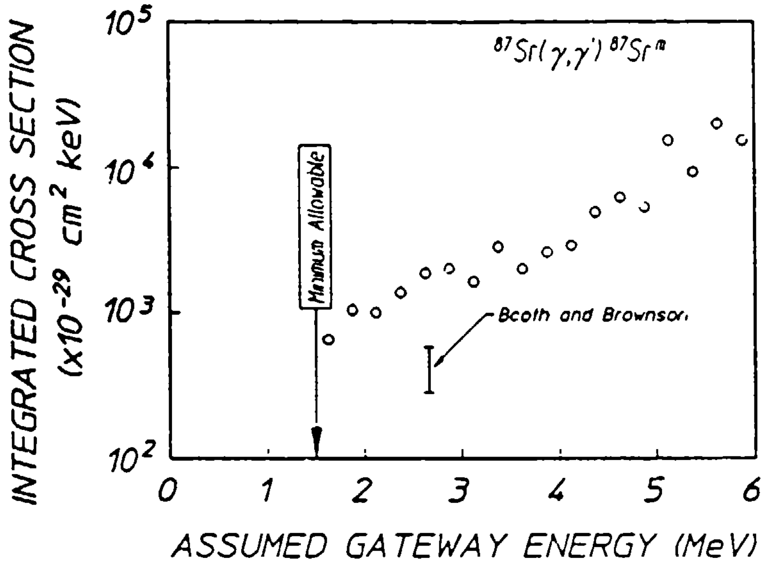
found between successive measurements at those times.[13] However, since our results are greater, an alternative explanation which relies on a complete agreement with the prior work demands the opening of additional gateways at higher energies.

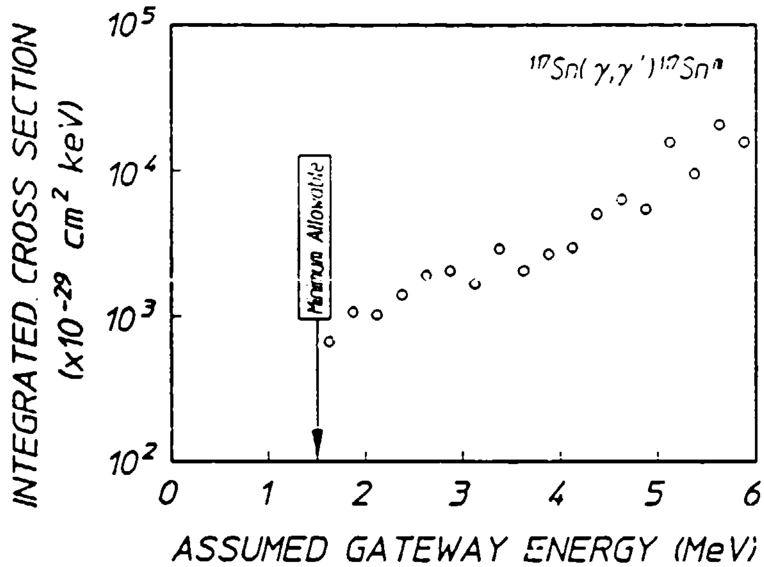
The pervasiveness found for the unexpectedly large values for integrated cross sections for the transfers of such large amounts of angular momenta suggests some type of core property varying only slowly with increasing nuclear size. In such a case, however, there would seem to be the need for a mixing of several single particle states so the decay of the gateway state could occur into several different cascades with comparable probabilities. In any case, the integrated cross sections found in this experiment correspond to remarkably large partial widths. Derived values are summarized in Table II. Such widths are characteristic of relatively unhindered E1 transitions and motivate further investigation of their occurrence.

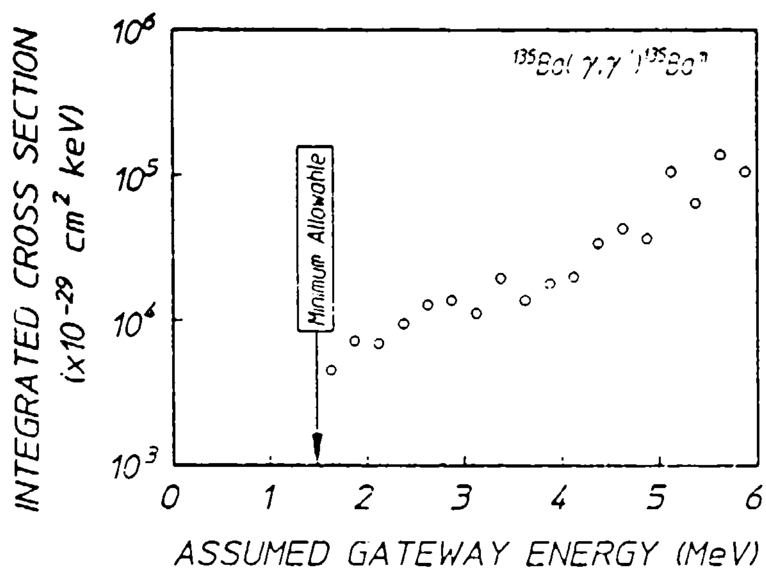
Table II

Summary of the partial widths for the (γ,γ') reactions producing the isomers of the species shown. Values were obtained from the integrated cross sections by assuming the gateway energies lay at 2 MeV, near the bremsstrahlung maximum, and that statistical weights were the same in the ground and gateway states, so that $\sigma_o = 6.1 \times 10^{-22}$ cm^2.

Nuclide	$\pi\sigma_o(b_a b_o \Gamma)/2$ at 2 MeV ($\times 10^{-29}$ cm^2 keV)	Partial width (meV)
^{87}Sr	1,000	10
^{117}Sn	1,030	11
^{135}Ba	6,900	72
^{195}Pt	22,900	240
^{199}Hg	1,740	18


INTEGRATED CROSS SECTION (x10^-29 cm^2 keV)
ASSUMED GATEWAY ENERGY (MeV)
87Sr(γ,γ')87Sr^m
Minimum Allowable
Booth and Brownson


INTEGRATED CROSS SECTION (x10^-29 cm^2 keV)
ASSUMED GATEWAY ENERGY (MeV)
117Sn(γ,γ')117Sn^m
Minimum Allowable


INTEGRATED CROSS SECTION (x10^-29 cm^2 keV)
ASSUMED GATEWAY ENERGY (MeV)
135Ba(γ,γ')135Ba^m
Minimum Allowable

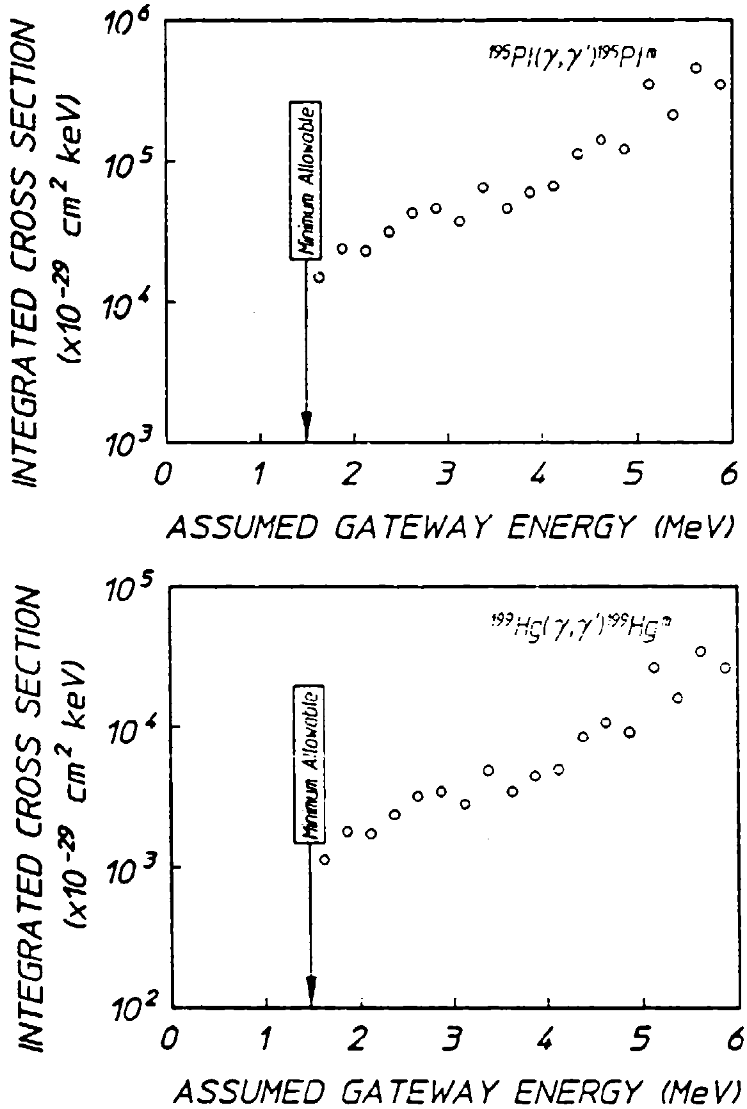


Figure 5: The integrated cross sections for the (γ,γ') reactions shown on each panel through single, unknown gateway states as functions of the energies at which they could be assumed to lie. Literature values preclude the possibilities that these gateways could lie at energies below the minima shown.
a) (opposite top) Reaction of ^{87}Sr together with a previous measurement from Ref. 8.
b) and c) (opposite center and bottom) Reactions of ^{117}Sn and ^{135}Ba.
d) and e) (top and center) Reactions of ^{195}Pt and ^{199}Hg.

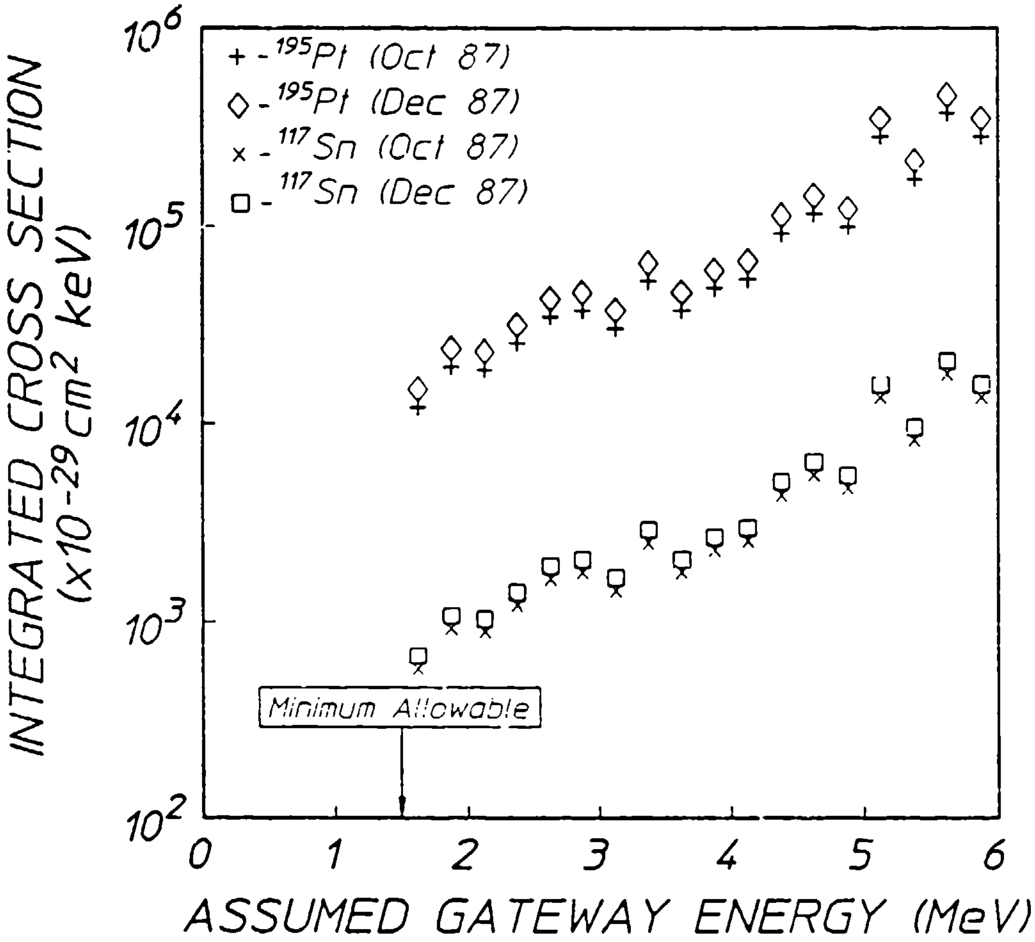


Figure 6: The integrated cross section for the (γ,γ') reactions producing the isomers $^{195}Pt^m$ and $^{117}Sn^m$ through single, unknown gateway states as functions of the energies at which they could be assumed to lie. Data from two separate experimental series are shown to illustrate long-term reproducibility.

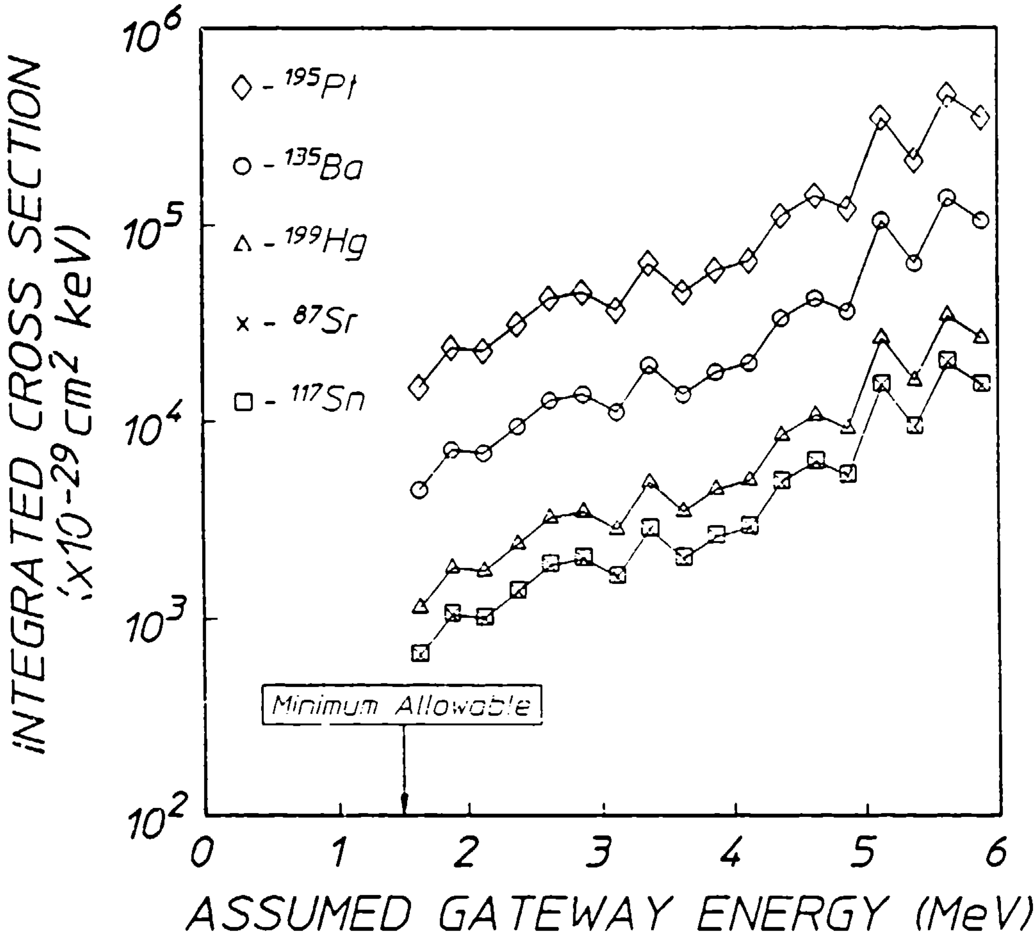


Figure 7: Summary of the integrated cross sections for the (γ,γ') reactions producing the isomers of the species shown, plotted as functions of the energies at which a single gateway state could be assumed for each.

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13. Difficulties in characterizing the source spectra led to wide variations in reported cross sections as reviewed in Refs. 5 and 6.

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